UNIVERSIDADE DE SANTIAGO DE COMPOSTELA

TESIS DOCTORAL

Valorización de lodos de depuradora como fertilizante en el marco de la economía circular: de residuo a recurso

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Dra. Dña. Mª Rosa Mosquera Losada, Dr. D. Antonio Rigueiro Rodríguez, Dra. Dña. Nuria Ferreiro Domínguez como directores de la Tesis Doctoral: **Valorización de lodos de depuradora como fertilizante en el marco de la economía circular: de residuo a recurso**

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Autorizamos:

La presentación de la citada Tesis Doctoral, realizada por Alberto Amador García dado que consideramos que reúne las condiciones necesarias para su defensa.

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Fdo. Mª Rosa Mosquera Losada

Fdo. Antonio Rigueiro Rodríguez

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Alberto Amador García

RESUMEN

La escasez futura de fertilizantes minerales y el aumento de sus costes económicos y ambientales, son razones clave para promover el uso de los residuos orgánicos biodegradables, entre ellos los lodos de depuradora, como fertilizantes en la agricultura. Además la producción de lodos de depuradora se ha incrementado en los últimos años con la obligación de depurar las aguas residuales desde 2005 en Europa. Por ello, es importante tener en cuenta los metales pesados presentes en los mismos, comparando las implicaciones de los valores límite actuales y el análisis de las implicaciones de establecer límites más estrictos por la Unión Europea (EU), para evaluar el efecto sobre la disponibilidad de sitios para el uso agrícola de lodos en Galicia. Los resultados indican que más del 90% de los suelos gallegos analizados son adecuados para recibir lodos con la actual normativa R.D. 1310/90, pero menos del 30% cumplirían con el último borrador de modificación de la normativa europea. Posteriormente es necesario valorar el comportamiento de la aplicación de lodos en los suelos disponibles, teniendo en cuenta el metal pesado regulado con la mayor concentración de lodos de depuración, que es el zinc, con diferentes escenarios de regulación, aplicación, dosis y tipo de lodo, para satisfacer las necesidades del cultivo. Resaltando la conveniencia de utilizar compost como enmienda, y el lodo granulado y el digerido anaeróbicamente como fertilizantes, para reducir la posible contaminación del suelo. En 2015, se aprobó el Paquete sobre Economía Circular para la UE, el cual propone objetivos sobre los residuos orgánicos, con el fin de poder usarlos como materias primas para la elaboración de abonos y crear un mercado regulado para su uso en la UE, reduciendo con ello los residuos, el consumo de energía y los daños ambientales. Por ello, es importante caracterizar el efecto de estos abonos o enmiendas orgánicas, elaborados con lodos de depuradora estabilizados con cal, sobre la fertilidad del suelo y el crecimiento de pastizales. Pudiendo ser una alternativa viable para suplir las necesidades de cal en los pastos y otros cultivos, e incluso parcialmente de fertilizantes minerales, reduciendo los costes de producción en las explotaciones, y el impacto ambiental de los residuos y los fertilizantes químicos.

ÍNDICE GENERAL

INTRODUCCIÓN

1. Necesidad de reciclaje de residuos

Los seres humanos han provocado la degradación de los suelos, agua y el territorio desde tiempos inmemoriables (Zhao, 2013). La preocupación por el medio ambiente, que comenzó a finales del siglo pasado, obligó a tomar decisiones para utilizar el suelo y el agua de una manera más sostenible (Meshesha et al., 2012), aspectos que se pueden vincular en la actualidad a la estrategia europea de bioeconomía (CE, 2012) y economía circular (CE, 2015).

El concepto de bioeconomía se basa en abordar desafíos sociales interconectados como la seguridad alimentaria, la escasez de recursos naturales, la dependencia de los recursos fósiles y el cambio climático, al tiempo que se logra un crecimiento económico sostenible (CE 2012). La bioeconomía proporciona una base útil para la sostenibilidad, ya que pretende una producción que emplea recursos biológicos renovables y la conversión de estos recursos y flujos de desechos en productos de valor añadido, como alimentos, piensos, productos biológicos y bioenergía. Por lo tanto, en el marco de la sostenibilidad y de la bioeconomía se hace imprescindible añadir valor a los productos de desecho y residuos, de los cuales los lodos de depuradora son de los más importantes debido a las grandes cantidades que se generan en Europa y a escala global tal y como se reconoce en el documento de la FAO "Smart Climate Agriculture" (FAO 2013). Por ello, en el año 2015 (CE 2015), la Comisión Europea adoptó un ambicioso paquete de medidas sobre la economía circular (Figura 1), que incluye propuestas legislativas para estimular la transición de Europa hacia una economía circular que impulsará la competitividad global, fomentará el crecimiento económico sostenible y generará nuevos empleos.

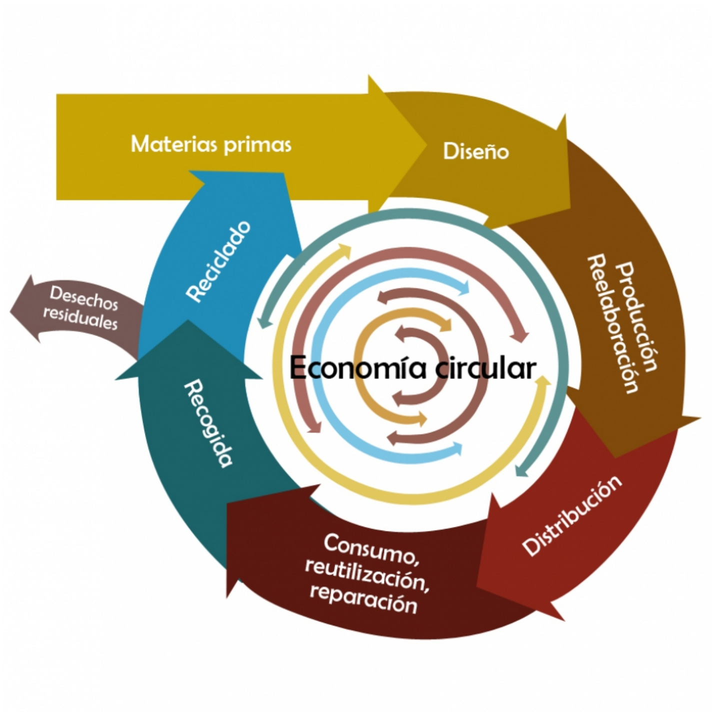

Figura 1. Esquema sobre la Economía Circular (EOI, 2017).

El paquete de medidas vinculado a la bioeconomía se enmarca en el plan de acción de Economía Circular de la Unión Europea (EU) y abarca todo el ciclo de vida y cadena de valor de los productos: desde el diseño, abastecimiento, la producción y el consumo hasta la gestión de residuos y el mercado de materias primas secundarias. En materia de residuos este plan de acción busca el llamado "closing the loop" (cierre del círculo) de los ciclos de vida de productos a través de una mayor reciclaje y reutilización, y para así aportar beneficios tanto para el medio ambiente como para la economía.

En la UE, la generación de residuos, excluidos los grandes residuos minerales (2004-2012), se distribuyó entre un 4,3% de aquellos provenientes de la agricultura, la silvicultura y la pesca y un 19,6% de las aguas residuales (Eurostat, 2016), La valorización de residuos orgánicos como enmiendas para su uso agrícola es una forma eficaz de reciclar la materia orgánica y los nutrientes que contienen (Stevens 2011;

Scotti et al., 2015, S et al., 2016). Por ello, una de las principales propuestas de la Comisión Europea dentro de su Paquete de Economía Circular (CE, 2015) es una normativa común sobre fertilizantes vinculada a la valorización de residuos. Esta normativa pretende estandarizar los diferentes documentos legislativos estatales que en la actualidad obstaculizan la fabricación y comercialización de fertilizantes a partir de residuos orgánicos, como por ejemplo de desecho de alimentos, los lodos de depuradora de la agroindustria o de estiércol. Es por ello que en el año 2016, la Comisión Europea puso en marcha la revisión de la reglamentación de la Unión Europea sobre los fertilizantes para ayudar a desarrollar un mercado de bio-nutrientes en la UE garantizando la seguridad y la calidad de este tipo de fertilizantes en la UE (CE, 2016). Esta normativa no incluye inicialmente los lodos de depuradora urbana por la elevada carga contaminante que poseen (metales pesados, compuestos orgánicos no deseados), pero si incluye los desechos procedentes de industrias agroalimentarias.

2. Generación de lodos de depuradora urbana y gestión

Los altos niveles de contaminación en aguas continentales promovieron en Europa el establecimiento de la directiva 91/271/CEE de aguas residuales en los años noventa. Esta directiva que mejoró la calidad de aguas continentales y costeras, provocó un aumento de la producción de lodos de depuradora urbana en Europa desde los años 90 pasando desde una producción anual de alrededor de 5,5 millones de Mg de materia seca (MS) en 1992 hasta casi 9,8 millones de Mg en 2012 (Eurostat, 2016). Estos residuos requieren una gestión adecuada y han provocado que las aguas residuales y los lodos se empezaran a utilizar como fuente de nutrientes en muchos países (Ghafoor et al., 2012).

La gestión adecuada de los lodos de depuradora se puede hacer por depósito en (i) vertedero (limitado por la actual normativa sobre vertederos, Real Decreto 1481/2001 (BOE, 2002), que condiciona el vertido a los contenidos de materia orgánica del lodo (como mínimo la cantidad total (en peso) de materia orgánica en los residuos urbanos biodegradables destinados a vertedero no puede superar el 35 por 100), por (ii) incineración que causa la liberación de nitrógeno y otros compuestos a la atmósfera a través de su volatilización durante el proceso de quemado del lodo (Smith, 1996; EPA, 2012) o (iii) mediante su uso en suelos como abono o enmendante. El uso de lodos de depuradora como fertilizante o enmendante en los suelos es promovido por la Unión Europea debido a los numerosos trabajos científicos que abogan por su empleo con este fin con base en su elevado contenido en nutrientes, principalmente nitrógeno, pero también en fósforo y materia orgánica, lo que podría aumentar el valor de este residuo que debe considerarse como un subproducto que de otro modo generaría problemas ambientales (Smith, 1996; CEE, 91; Mosquera-Losada et al., 2011b). Si el lodo se gestiona adecuadamente, su uso mejora la fertilidad del suelo en nutrientes y aumenta la materia orgánica del mismo (BOE, 1990; EPA, 1994; Sigua et al., 2005; Mosquera-Losada et al., 2010), lo que está en consonancia con la promoción de sistemas agrarios en el marco de la bioeconomía y economía circular, proporcionando un valor al residuo, convertido en recurso. Una de las principales preocupaciones del uso de lodos de depuradora como fertilizante en suelos está relacionada con el hecho de que el lodo presenta unos niveles más altos de metales pesados y otras sustancias orgánicas no deseables que los suelos, que podrían ser absorbidos por las plantas y afectar a los seres humanos a través de la cadena trófica (Roy y McDonald, 2013). Además, el uso de lodos de depuradora como fertilizante podría causar una degradación importante del suelo, incluyendo daños físicos, químicos y microbiológicos (Yang et al., 2012), lo que podría tener un impacto a corto, medio y largo plazo y por lo tanto afectar la salud del mismo (Fernández- Calviño et al., 2013) si su empleo no se realiza de forma adecuada.

El empleo de lodo fresco en agricultura no está permitido en Europa, es por ello que deben estabilizarse para evitar la presencia de patógenos, algo que sí ocurre en otros países como Estados Unidos. El uso de lodos de depuradora en agricultura ha de realizarse con lodos tratados y estabilizados, de manera que reduzcan su poder de fermentación (olores) y mejoren su calidad microbiológica por higienización (BOE, 1990; EC, 2001). Este proceso de estabilización de los lodos de depuradora modifica su valor fertilizante y por lo tanto la dosis de aplicación para satisfacer las necesidades de los cultivos (EPA, 1994; Mosquera-Losada et al 2010). La forma de estabilización suele seleccionarse en función del volumen de la producción de lodos de depuradora (Mosquera-Losada et al., 2010). Los tratamientos más comunes y promovidos de lodos de depuradora para reducir la contaminación microbiana y olores así como favorecer el uso agronómico son la digestión anaeróbica y el compostaje, con un tiempo de tratamiento corto y largo, respectivamente. Después de los tratamientos anaeróbicos, la deshidratación por tratamiento térmico de los lodos de depuradora es útil cuando se persigue la reducción del volumen del lodo almacenado y los costes de transporte asociados a su uso como fertilizante o enmienda, facilitando al mismo tiempo su aplicación al suelo mediante el empleo de maquinaria que existe actualmente en las explotaciones agrarias o en las empresas de servicios agrarios. Otros tratamientos avanzados son la estabilización con cal, que basa su efectividad en elevar el pH del residuo hasta un pH en donde el desarrollo microbiano está muy limitado (EPA, 1994;

EC, 2001). La estabilización alcalina con cal es un tratamiento avanzado (EC, 2001) para residuos orgánicos que se considera una alternativa atractiva a la digestión aerobia o anaeróbia (Czechowski et al., 2006), aunque hoy en día su uso es minoritario en la UE (Werle et al., 2010) de gran importancia en Galicia por la acidez de sus suelos, aspecto que se evalúa en esta tesis.

Por la presencia de metales pesados, patógenos y compuestos orgánicos no deseables en el lodo que es aplicado al suelo que pueden causar daño al entorno o a la población, la Comisión Europea estableció limitaciones vinculados a una serie de metales pesados para el uso de lodos de depuradora en el año 1986 (UE, 1986), desarrolló un borrador de normativa en el año 2000 (EC, 2000), 2008 contrató la realización de un informe sobre varios escenarios normativos alternativos (EC, 2008) en el año 2008 y la elaboración de un informe al *Join Research Centre* de Ispra, Italia (JRC, 2012). Derivada de la normativa establecida en el año 1986 y que se traspuso en España en el Real Decreto 1310/90 en la actualidad existen tres tipos de condicionantes que regulan el uso de lodos de depuradora en agricultura en base al contenido en metales pesados: (i) valores máximos de concentración en el lodo, (ii) valores máximos de concentración en el suelo y (iii) cantidad máxima media de metal pesado que puede aplicarse durante un período de diez años en kg por hectárea y año. Sin embargo, existe un acuerdo entre los responsables políticos, los científicos y la población de que los umbrales de metales pesados deben limitarse aún más modificando la Directiva de la UE 86/278 / CEE (UE, 1986). Por esta razón, la Comisión Europea presentó un proyecto de documento de trabajo en 2000 (CE, 2000), que todavía está en discusión y que no se implementó finalmente debido a los costes y a los problemas relacionados con la proposición de límites restrictivos que algunos países no eran capaces de cumplir. La

Comisión Europea (CE, 2008) en el documento "Environmental, economic and social impacts of the use of sewage sludge on land. Final Report" ha evaluado estos límites y planteado varios escenarios empleando límites con diferentes grados de restricción que el anteriormente mencionado (EC, 2000), evaluando las ventajas e inconvenientes que tendría en su implementación. En ambos proyectos (CE 2000 y CE 2008), la aplicación de lodos de depuradora estaría prohibida en suelos con pH inferior a 5, aspecto relacionado con la mayor disponibilidad de la mayoría de los metales pesados a pH ácidos. En todo caso, la producción de lodos de depuradora de alta calidad es el primer aspecto que debe abordarse para utilizar un residuo de forma sostenible mejorando la fertilidad del suelo, tal y como ya señalaba la Comisión Europea (EC, 2000). La evaluación de los suelos es muy relevante porque los niveles de pH y concentración de metales pesados en el suelo son esenciales para evaluar las necesidades de fertilidad del mismo así como el riesgo de la incorporación de lodos al suelo y la posibilidad de que los metales pesados lleguen a los seres humanos causando problemas de salud como cáncer (Tchounwou et al., 2012). Esta evaluación es más relevante en aquellas regiones con alta demanda de fertilizante debido a sus buenas condiciones ambientales (temperatura, humedad, radiación) y con suelos ácidos debido al hecho de que los metales pesados están más disponibles en suelos ácidos (Porta et al, 1999).

3. El suelo y el empleo de lodos de depuradora como fertilizante en Galicia

En Galicia (NW España), la industria alimentaria y las actividades primarias (agricultura, silvicultura y pesca) y agua/residuos representan más del 11% del producto interno bruto (PIB). El sector primario representó más del 18% del total de industrias en

Galicia en 2014. En esta región, en el año 2014, se generaron 2,2 millones de toneladas de Residuos No Peligrosos (RNP) a partir de actividades industriales y el 82% se recicló y se reutilizó (Xunta de Galicia, 2016) en operaciones de recuperación como las establecidas en el Anexo II De la normativa española y comunitaria sobre residuos (EC, 2008, BOE, 2011). Aproximadamente el 25,8% del total de RNP distribuidos en lodos de depuradora (8,5%), biorresiduos (8,7%), cenizas de madera (7,8%) y estiércol %) tienen el potencial, como residuo orgánico, de ser utilizados como materia prima para el desarrollo de fertilizantes o sustratos.

Esta región española, La Comunidad Autónoma de Galicia, que ocupa alrededor de 3 millones de hectáreas (IGE, 2011), produce el 53% de la madera en España (datos de 2012) (INE, 2015), pero también genera el 34% de la oferta española de leche con base forrajera (cultivos como el maíz) y pastos (INE, 2015). Los terrenos de cultivo, de pastos y forestal representan el 12,9%, el 15,1% y el 60,9% de la superficie total de Galicia, respectivamente (IGE, 2016). La importancia del sector ganadero en Galicia hace imprescindible el cambio de sistemas intensivos a sistemas que empleen el pasto como base de la gestión ganadera con el objeto de abaratar los costes de producción vinculados a la alimentación. La elevada proporción de terrenos abandonados en Galicia cubiertos por matorral hace que en la actualidad la transformación en pastos sea de gran relevancia con el objeto de incrementar la sostenibilidad de los sistemas ganaderos en Galicia. Además en la actualidad, el 97% de los lodos de depuradora producidos en la región (188.384 Mg en 2014) se valorizan como fertilizante o enmendante del suelo (Xunta de Galicia, 2016).

Los suelos gallegos son ácidos y podrían considerarse representativos de una gran parte de la región biogeográfica atlántica de Europa (EC, 2005, AEMA, 2003, Clea et al. 2014). Debido a su acidez y a su gran potencial para la producción agronómica y ganadera presenta una gran necesidad de fertilizantes y enmiendas. En Galicia, la acidez del suelo es un fenómeno natural derivado principalmente de su clima húmedo, con una mayor precipitación que la evapotranspiración durante la mayor parte del año (Álvarez et al., 2009), lo que conlleva un mayor lavado, una merma del nivel de cationes en el suelo y la consiguiente disminución de su fertilidad que se asocia a un exceso de Aluminio en el mismo (Mombiela y Mateo 1984; López-Mosquera 1995). Por lo tanto, es aconsejable realizar actividades de manejo como el encalado y la fertilización para aumentar la fertilidad del suelo y neutralizar la acidez que puede limitar la producción de los cultivos. Es por ello que el aporte de enmiendas orgánicas, como los lodos de depuradora urbana con pH alrededor de 7 o muy superiores en el caso de que se sometan a procesos de e estabilización con cal viva, que suplementa el efecto fertilizante y el aporte de materia orgánica y nutrientes de los residuos orgánicos, es de gran interés.

4. Empleo de lodos como fertilizante y su efecto en el suelo y pasto

4.1. Propiedades físicas y químicas del suelo

La fertilización orgánica resulta ser menos eficiente a corto plazo que en el caso de la fertilización mediante abonos químicos de síntesis, que por el contrario causan grandes problemas medio ambientales y descapitalizan el suelo en términos de materia orgánica y micronutrientes. Esto se debe a que el fertilizante orgánico debe previamente mineralizarse para liberar los nutrientes, siendo este un proceso lento y variable según las condiciones existentes en el suelo, que intervienen en el desarrollo de la población microbiana encargada de esta mineralización, como son la temperatura, el contenido en humedad o el pH (EPA, 1994; Rigueiro-Rodríguez et al., 2010b; Mosquera-Losada et al., 2012; Ferreiro-Domínguez et al., 2011; Ferreiro-Domínguez , 2011).

En general, la aplicación de lodo afecta a las propiedades físicas y químicas del suelo. En el caso de las propiedades físicas, este tipo de residuos mejora la estructura y la estabilidad de los agregados, gracias al aporte de materia orgánica que se realiza, lo que da lugar a un incremento en la permeabilidad y en la retención hídrica (Navarro-Pedreño, 1995).

En cuanto a las químicas, sobre el pH del suelo depende del tipo de lodo que se emplee y del suelo en el que se aplique, en suelos muy ácidos estudio anteriores muestran un incremento del pH debido al aporte de cationes que se realiza, en especial el calcio (López-Díaz et al., 2007; Rigueiro *et al*., 2002a). En suelos con pH alto limitan la reducción de pH en suelos con tendencia a la acidez como es el caso de los gallegos (Mosquera-Losada et al., 2006; Rigueiro-Rodríguez et al., 2012b). Este incremento del pH sobre suelos ácidos reduce la presencia de aluminio en el complejo de cambio, debido al incremento de calcio que se producen con el lodo de depuradora, tal y como encontraron Vivekanandan et al., (1991) y López-Díaz (2004), y también incrementa la disponibilidad de Mg (Vivekanandan et al., 1991; López-Mosquera et al., 2002). Este efecto es más evidente en aquellos lodos que han sido estabilizados con cal y que, por tanto, presentan mayores contenidos de Ca (Rigueiro *et al*., 2002b; Rigueiro *et al*., 2004). Sobre el contenido en materia orgánica, su incremento se hace más evidente con sucesivas aplicaciones de lodo al suelo (Tsadilas et al., 1995; Krebs et al., 1998) o con el aumento de las dosis aplicadas (Canet et al., 1996; Lindsay *et al*., 1998). Sin embargo, también existen suelos en los que esto no ocurre, debido a la rápida mineralización del lodo o a que el suelo al que se aporta posee una elevada proporción de materia orgánica en comparación a la aplicada con el lodo (Gigliotti et al., 2001). En cuanto a la capacidad de intercambio catiónico del un suelo puede verse mejorada tras el aporte de lodo, sobre todo si se incrementa el contenido de materia orgánica (Piccolo et al., 1992). En cuanto a los macronutrientes, la aplicación de lodo de depuradora urbana al suelo produce un incremento de los niveles de nitrógeno en el mismo (Rodríguez-Barreira, 2007), lo que se relaciona con el aumento de materia orgánica y su mineralización) y que se refleja en un incremento de la producción. Otros estudios demuestran un aumento de la presencia de fósforo total, principalmente cuando existe una carencia del mismo (Mosquera-Losada et al., 2008) y del fósforo intercambiable (Cucci et al., 2008). En cuanto al potasio, suele aparecer en el lodo en formas fácilmente asimilable por las plantas (Sabey y Hart, 1975), por lo que el aporte de lodo de depuradora mejora su disponibilidad (O´Riordan *et al*., 1987), incluso aquellos que han sido estabilizados con cal, a pesar del antagonismo existente entre el K y el Ca (Vivekanandan *et al*., 1991). Finalmente, la cantidad de sodio también suele incrementarse debido a la descomposición de estos residuos (López-Mosquera et al., 2002).

La importancia de los suelos ácidos en Galicia, la gran cantidad de suelos no empleados destinado a matorral y el gran potencial ganadero que ha de reconvertirse de sistemas intensivos a aquellos que empleen más pastos con el objeto de mejorar su rentabilidad ha sido la base para evaluar el efecto del aporte de lodos de depuradora urbana sobre la mejora edáfica en suelos de monte muy ácidos para su conversión en pastos en esta tesis.

4.2. Metales pesados

El efecto del lodo de depuradora sobre los niveles de metales pesados en el suelo va a depender de las dosis aportadas, cuyos máximos están regulados por el RD 1310/90, la concentración de metales pesados en el suelo (además del pH) y fango y finalmente del potencial contaminante de los metales pesados.

Los niveles de metales pesados en el suelo, son de gran importancia ya que si un lodo de depuradora tiene unos niveles de metales pesados muy inferiores a los marcados por la normativa vigente (RD 1310/90), por tanto pudiendo caracterizarse como un lodo de gran calidad, pero el suelo al que se pretende aportar tiene unos niveles de metales pesados muy altos, aunque solo sea uno, el lodo no podrá emplearse como fertilizante. Es por ello que, en este estudio, y gracias a la colaboración con la empresa AGROAMB hemos realizado una evaluación del nivel de los metales pesados regulados en numerosas fincas a las que no se han aportado lodos de depuradora con el objeto de establecer el nivel base de metales pesados en suelos agrícolas de Galicia.

Si nos centramos en la concentración de metales pesados en el lodo regulada en la actualidad (cromo, zinc, cobre, mercurio, cadmio, níquel y plomo) para el empleo de lodos de depuradora, este residuo suele tener elevados niveles de zinc, seguido del cobre y cromo (Smith, 1996; Mosquera-et al. 2009c, 2010). Es por ello que en este estudio realizamos un análisis del empleo de los lodos de depuradora urbana en suelo centrándonos en el Zn ya que se convierte en la actualidad en el primer limitante para el el empleo de lodos de depuradora urbana en agricultura en Galicia a corto plazo.

El Zn y el Cu suelen existir en gran cantidad en las aguas residuales contaminadas por las industrias. Estos elementos son esenciales para la planta (Zn y Cu) y para el animal (Zn, Cu y Cr), pero los demás (Hg, Cd, Ni y Pb) no lo son, lo que les confiere un mayor riesgo de toxicidad. De todos los elementos regulados, el cadmio es el que posee una mayor potencialidad toxicológica para las personas que consumen productos procedentes de suelos tratados con lodos residuales, por ser fácilmente absorbido por las plantas. Afortunadamente, el plomo y el cromo, que son los principales contaminantes de ciertas zonas, no son absorbidos con tanta facilidad por las plantas como el cadmio, pero sus niveles deben ser estrictamente controlados. En concreto, la disponibilidad del plomo es reducida debido a que este elemento se encuentra en los horizontes superficiales ligado a la materia orgánica del suelo (Canet et al., 1998), y prácticamente no se lava (Davies, 1980). Todos estos elementos poseen una persistencia prolongada en el suelo y pueden pasar bastantes años hasta que desaparezcan sus efectos tóxicos (Simpson, 1986; Pomares y Canet, 2001). En este estudio, no se analizan estos aspectos, ya que nos centramos en el Zinc como metal pesado dominante, por lo que sus conclusiones en relación al lodo de depuradora deben tomarse con precaución desde un punto de vista holístico, ya que el mismo ejercicio debe realizarse con el resto de metales. Además, deben considerarse las modificaciones que desde un punto de vista de contenidos en sustancias orgánicas del residuo provoca el aporte de lodos de depuradora en agricultura. De hecho, la adición de lodo durante períodos de tiempo prolongados provoca aumentos en el suelo del contenido de cromo (Kabata-Pendías y Pendías, 2001), zinc, cobre y plomo (Andrade-Couce et al., 1985; Mosquera-Losada et al., 2010, 2009a), cadmio (Canet et al., 1998) y níquel (Canet et al., 1996; López-Díaz, 2004; Rodríguez- Barreira, 2007), dependiendo de las dosis aportadas y del número de aplicaciones. La mayoría de los metales pesados se acumulan a la profundidad a la que se ha incorporado el lodo, como es el caso del Cr (Berti y Jacobs, 1998; Canet et al., 1998), Cu (Barbarick *et al*., 1998; Berti y Jacobs, 1998; Canet *et al*., 1998), Zn (Berti y Jacobs, 1998), Ni (Alloway 1995; Berti y Jacobs, 1998) y Cd (Barbarick *et a*l., 1998; Berti y Jacobs, 1998; Canet *et al*., 1998).

4.3. Pasto

4.3.1. Producción de pasto

Existen diversas investigaciones que estudian el efecto de la fertilización con lodo sobre la producción de pasto en Galicia (Mosquera-Losada et al., 2006; López- Díaz et al., 2007; Rigueiro-Rodríguez et al., 2008) y en otras partes del mundo (Sibbald et al., 2001; Etienne, 2005; Pontes et al., 2007). En general, el aporte de lodo de depuradora mejora la producción de pasto, al igual que el abonado mineral si se ajustan las dosis de forma adecuada (Rigueiro-Rodríguez et al., 2010a).

Generalmente, la fertilización con lodo no estabilizados mediante el aporte de cal favorece la producción de pasto en suelos agrícolas (Mosquera-Losada et al., 2006), y más, si la fertilización con lodo de depuradora es acompañada de encalado, tal como encontraron Mosquera-Losada et al (2009). Es por ello que el aporte de lodos estabilizados con cal pueden tener un gran interés en suelos ácidos como los gallegos.En la producción de pasto influye también el tipo de estabilización del lodo utilizado para su abonado. De hecho, el aporte de lodo digerido anaeróbicamente y peletizado tiene un mejor efecto sobre la producción de pasto a corto plazo en comparación con el compostado, que por el contrario posee un mayor efecto residual de incremento de fertilidad en el suelo, debido a que generalmente presenta una tasa de mineralización inferior al resto (Rigueiro-Rodríguez et al. 2010b, Mosquera-Losada et al., 2016).

4.3.2. Biodiversidad

El efecto de la aplicación de lodo de depuradora sobre la calidad de pasto va a depender de las dosis de lodo aportadas y de la presencia de especies sembradas en el pasto (López-Díaz et al., 2007). La utilización de especies herbáceas de calidad, acompañadas de encalado y fertilización que mejorarán la fertilidad del suelo y por tanto favorecerán su desarrollo, son prácticas que permiten la mejora en la productividad y calidad del pasto (Piñeiro y Pérez, 1988; Silva-Pando et al., 1998).

Cabe señalar que la mejora de la fertilidad del suelo reduce la invasión de malas hierbas y por lo tanto la biodiversidad, tal como lo encontraron autores como Thompson et al. (2001) o Dise and Stevens (2005); o en el caso de la fertilización orgánica con lodos de depuradora, tal como encontraron autores como Mosquera-Losada et al. (2009), dónde también hallaron un efecto negativo en la alpha-biodiversidad cuando se combinaba el encalado con la fertilización con lodos de depuradora, al igual que Ferreiro-Domínguez et al. (2011). Estos impactos sobre la biodiversidad del aporte de fertilizantes orgánicos o inorgánicos dependen de la dosis de aplicación.

El efecto del aporte del lodo sobre la biodiversidad modifica las especies dominantes además del número de especies presentes, asociándose un mayor aporte de lodo al incremento de las especies gramíneas de siembra (Mosquera et al., 2001; López-Díaz, 2004), que son más exigentes en fertilidad edáfica, en comparación con el encalado que mejora la aparición de especies graminoides asociadas a zona de baja fertilidad nitrogenada como es el caso de las del género Agrostis, con lo que se mejora la calidad y productividad del pasto (Mosquera-Losada, 1999, 2001)). En general el aumento de especies de las familias del grupo de las leguminosas o dicotiledóneas se suelen asociar a dosis muy reducidas de abonos inorgánicos o orgánicos y al aporte de cationes como el potasio, que suelen presentar en mayores concentraciones de gramíneas (Mosquera-Losada et al., 2001b; López-Díaz, 2004, López-Díaz et al. 2007).

4.3.3. Contenido en metales pesados

La aplicación de lodos de depuradora como fertilizante modifica los niveles de metales en el pasto como consecuencia del incremento que provoca en el suelo, aspecto contemplado en esta tesis en la transformación de zonas arbustivas en pastos solo para el caso del zinc y que ha sido previamente estudiado en Galicia en sistemas silvopastorales (López-Díaz, et al., 2007; Mosquera-Losada et al., 2001a, 2009a). En diferentes estudios se ha visto que el aporte de lodo incrementa en pasto las concentraciones de cobre (Mosquera-Losada et al., 2001a; Tiffany et al., 2000b; Mosquera-Losada et al., 2009a), de zinc (Loué, 1988; Mosquera-Losada et al., 2009a), de Mn (Williams et al., 1997) y de cadmio (Tsadillas et al., 1995). Sin embargo, no se ha encontrado que la aplicación del lodo de depuradora al suelo provoque modificaciones en la concentración de cromo (Hamon et al., 1999; López-Díaz, 2004) y plomo (Canet et al., 1998; López- Díaz, 2004) en el pasto en los estudios evaluados. En cuanto al contenido de níquel en planta, éste depende de la disponibilidad de níquel en el suelo; así, Sanders et al. (1986) observó un incremento de este elemento en la planta al emplear este tipo de residuos en un suelo básico en el que se produjo una reducción de pH. Del mismo modo en Galicia López-Díaz (2004) observó un incremento de níquel en planta proporcional a la dosis de fertilizante orgánico empleado cuando el pasto crecía sobre un suelo ácido.

OBJETIVOS

Por ello, el **objetivo** de la tesis es la evaluación y análisis del empleo potencial de lodos de depuradora urbana en Galicia con base en el análisis del nivel base de metales pesados en los suelos gallegos (capítulo 1), el desarrollo de escenarios para evaluar el efecto del aporte de Zn con el lodo considerando los niveles base de suelo y la normativa vigente (capítulo 2) y el estudio sobre el efecto del aporte de lodos cuando son usados como materia prima en la elaboración de enmiendas orgánicas, en la transformación de montes en pasto a través de la evaluación de la mejora de la fertilidad del suelo, la producción de pasto y su biodiversidad (capítulo 3).

RESULTADOS

Capítulo 1. Sustainable use of sewage sludge in acid soils within a circular economy perspective

Abstract

Fertiliser future shortage and the associated increased economic and environment transport costs are key reasons to promote the use of urban residues as fertilisers in agriculture. Sewage sludge (SS) use as a fertiliser is promoted by the EU, which also consider the harmful effects of heavy metals (HMs). It is important to characterise the levels of HM in soils allocated to pasture and forage crops, before SS application, in order to have an initial soil reference and to see how they vary over time specially in acid soils, where HM availability and therefore its ecosystem im- pact is larger. For this study, we selected a region with natural very acid soils, and with high needs of fertilisers due to the high crop production potential regarding to important crops like maize, forage crops and grasslands. Galicia is a small region of Spain (9% of the Spanish territory) that produces the 33% and the 60% of the woodland products of Spain. This study generally aims to evaluate the use of SS as a fertiliser in a large agronomic region of Spain, as well as to identify the disadvantages associated with its use. In concrete terms, the study aims at comparing the implications of the current limit values and the analysis of the implications of tighter limits imposed by the EC to evaluate the effect on the availability of sites for sludge applications in Galicia. Results indicate that, after the analysis of 2557 soils, more than the 90% of Galician soils are suitable to receive sewage sludge (SS) following the current regulation RD 1310/90 but only 28.7% fulfil the EUDWD (European Union Draft Working Document) requirements. Most of the samples that do not fulfil the Spanish regulation are associated with basic and ultrabasic rocks that define natural environments with specific plants and soil microorganisms already adapted to these levels of HM. In order to apply more sustainable practices for agricultural production, it is proposed to take into account

the mean HM levels of the soil for each heavy metal (HM) trying not to surpass the mean levels of the soils derived from the different parent rock material, after considering human health risks. Moreover, this recommendation would respect the original environment of the soil that acts as a habitat for different organisms, pre-serving beta biodiversity.

1. Introduction

Soil sustainable use and health is one of the key aspects to maintain and promote sustainable agriculture production. Fertilization is one of the most spread management activity linked to agriculture. However, due to the economic and environment costs of the fertiliser transport, fertilization based on farm surrounding residues is promoted within the FAO mixed farming concept (FAO, 2015). Urban-agriculture ex- change of energy and nutrients should be promoted through activities (i.e. kilometer zero) like the use of urban residues in agricultural lands. The bioeconomy concept relies on addressing inter-connected societal challenges such as food security, natural resource scarcity, fossil resource dependence and climate change, while achieving sustainable economic growth. The bioeconomy concept provides a useful basis for such approach, as it encompasses the production of renewable biological resources and the conversion of these resources and waste streams into value added products, such as food, feed, bio-based products and bioenergy (EC, 2012). It is essential to add-value to waste products, of which SS is one of the most important ones due to the large amounts of production of this residue in Europe as already recognized the FAO Smart Climate Agriculture document (FAO, 2013).

High levels of contamination in continental waters caused by humans promoted the establishment of the waste water directive in the nineties in Europe. Waste water is being also used as a source of nutrients in many countries (Ghafoor et al., 2012). The production of municipal SS in Europe has been increased since the start of the 1990s of the last century due to the implementation of the 91/271/CEE Directive, which makes compulsory to treat continental waters in all cities with N 2000 inhabitants. So, the availability of this residue is ensured and usually perceived as a challenge due to the high level of nutrients that can provide to crops.

Sewage sludge elimination could be done by transport to landfill, by incineration, which causes nitrogen release into the atmosphere (Smith, 1996; EPA, 2012) or by using it in soils. The use of SS as a fertiliser in soils is promoted by the European Union due to its nutrient contents, mainly nitrogen but also phosphorus, which could increase the value of this residue that otherwise, would generate environmental problems (Smith, 1996; 91/271/CEE Directive). This practice is also in line with the bioeconomy and circular economy concepts, providing an added value to the residue. One of the main concerns of SS use in soils as a fertiliser is related to the higher levels of HM and other organic substances com- pared with soils, which could be absorbed by plants and affect human beings through the food chain (Roy and McDonald, in press). Moreover, the use of SS as fertiliser could cause important soil degradation, including physical, chemical and microbiological damages (Yang et al., 2012), which could have a short, medium and long term impact and therefore affecting soil health (Fernández-Calviño et al., 2013).

The European Directive 91/271/EEC which has the same HM thresholds as the Spanish Royal Decree 1310/1990 established limits for the use of SS as a fertiliser mainly by focusing on HMs. Moreover, the European Union launched a draft of the Directive to reduce even further the limits of allowable SS use in agriculture, on the European Union scale, in 2000 ((EC, 2000) (Brussels, 27 April 2000 - ENV. E.3/LM)).

The so called 3 draft working document on sludge produced by the EU (EUDWD (EC, 2000)) was not approved due to the lack of consensus between the different countries but it could give us an idea of the next steps that will be taken in the regulation of the use of SS in agriculture at a European level and how to become more eco-efficient. This confirms that it is not easy to establish more strict limits at European level, and that other approaches are needed. The first step to reduce the impact of SS use is to monitor soils previous SS inputs and control how the soil evolution will take place (Vacca et al., 2012).

The HM concentrations of the SS, of the soils where it would be ap- plied and the maximum quantity of HMs that could be applied in a 10 year period are currently limited by the approved regulations. The limits also depend on the soil pH, as HM availability increases as soil acidity is raised (Kabata-Pendias, 2001; Parat et al., 2005; Smith, 2009). Taking into account these directives, it is important to know what the base concentrations of HMs in municipal SS are, in different types of SS, that have already been evaluated at the Spanish national level (Mosquera-Losada et al., 2010). The second aspect to be evaluated is the level of HMs that already exist in the soils prior to adding SS, in order to study their capacity to receive SS as fertiliser under current and future directives, ensuring soil sustainability also for soil microorganisms (Guiller et al., 2009).

Galicia is a region with a large surface allocated to forestry (above 70% and producing 60% of wood in Spain (data from 2009)) (INE, 2012) but this Spanish region also produces N 33% of the Spanish milk supply (data from 2008) mainly based on forage (crops like maize) and grasslands (INE, 2012). The use of SS on forested areas in low doses (50–100 kg of total N ha^{-1}) has been successfully tested in the north of Spain (Egiarte et al., 2009; Mosquera-Losada et al., 2011a) as well as in higher doses (160 kg of total N ha^{-1}) on grasslands (Ferreiro-Domínguez et al., 2011; Mosquera-Losada et al., 2011a, 2011b; Rigueiro-Rodríguez et al., 2000b, 2010a, 2010b, 2012). The use of SS on these types of land reduces the possible negative impact of HMs on human beings, as there is no direct consumption of the plants that may take up the HMs from the soil. Currently, N 65% of SS produced in the region is used as fertiliser. Galicia soils could be considered repre- sentative of a large part of the Atlantic biogeographic region of Europe (EC, 2005; EEA, 2003). This study generally aims to evaluate the use of SS as a fertiliser in a large agronomic region of Spain, as well as to identify the disadvantages associated with its use. In concrete terms, the study aims at comparing the implications of the current limit values and the analysis of the implications of tighter limits imposed by the EC to evaluate the effect on the availability of sites for sludge applications in Galicia.

2. Materials and Methods

The study was carried out in Galicia, a region found in the northwest of Spain, which occupies around 3 million ha. It is within the southwest part of the Atlantic biogeographic region of Europe. From 2007 to 2010, 2597 soil samples, which were never previously fertilised with SS, were taken randomly from privately owned plots in order to see whether they were suitable to receive SS. Soil samples were taken mostly from the agrarian based counties of the Galician region, where more fertiliser is needed compared with the predominantly forested counties (Fig. 1). The main agrarian activity in Galicia is related to forage crops and pasture to feed dairy and meat cows (IGE, 2011). The main forest species are *Pinus pinaster* Ait. and *Eucalyptus globulus* Labill, followed by a mixture of each of these two species with *Quercus robur* L. Soil was sampled at a depth of 25 cm as established by the Spanish Royal Decree 1310/90. Each sample was taken following the procedure indicated by the Spanish Royal Decree 1310/90: "a representative sample of soil per plot sub- mitted to analysis will be composed of a mixture of 25 samples taken from an area of 5 hectares or less per plot". The number of hectares of the sampled plots represents approximately 20% and 1.5% of grasslands and agrarian soils (including forage crops and grasslands) of Galicia, respectively. Once taken, all soil samples were transported to the laboratory and air dried. Afterwards, soil samples were sieved through a 2 mm sieve. Later on, Ni, Cd, Zn, Cr, Cu and Pb concentrations were analysed with the VARIAN 220FS spectrophotometer using atomic ab- sorption (VARIAN, 1989), after a nitric acid digestion made in a CEM MDS-2000 microwave (CEM, 1994), with Hg determined with hydride generator. Water soil pH was also measured (2.5:1) (Guitián and Carballas, 1976).

Descriptive statistics (mean, median, kurtosis, etc.) of the HMs were performed on the 2597 samples in the experiment and counting the number of samples that meet or fail legal requirements for SS use in agriculture. The descriptive statistics include median absolute deviation (mad), which is a robust method for evaluating dispersion. The t Stu- dent and U Mann Whitney tests were used for comparison of HMs between samples that fulfil and those samples that do not fulfil the Spanish regulations. The obtained results with both tests were similar due to the large sample size in this comparison that tended to diminish the detrimental effects of non-normality. Factor analysis and cluster analysis were used to evaluate the relationships between the concentrations of different HMs in those soils that did not fulfil the current regulations. Using only the metals with significant differences between the samples that meet and fail the current legal requirements, a factor analysis from an exploratory perspective was performed. Factor analysis provides two outcomes; data summarisation and data reduction (Hair et al., 2010). In summarising the data, factor analysis derives the underlying dimensions that, when interpreted and understood, describe the data in a much smaller number of concepts than the original individual variables (HMs). Data reduction extends this process by deriving an empirical value (factor score) for each dimension (factor) and then substituting this value for the original values. The purpose is to simplify the subsequent multivariate analyses.

The analyses of the similarities and differences among those plots that do not fulfil the Spanish regulations were performed with a dendogram derived from a cluster analysis applied to the standardised HMs data. Proximities between samples were calculated based on the squared Euclidean distance and Ward's method was used as linkage procedure. The cluster analysis is complementary to the factor analysis. Factor analysis makes groupings based on the patterns of variation (correlation) in the data, whereas cluster analysis makes groupings on the basis of distance (proximity) (Hair et al., 2010). Statistical calculations were performed using SPSS (PASW 18.0) for windows.

3. Results

Table 1 shows the results of the descriptive statistics of soil pH and HMs obtained from the 2597 samples taken in this experiment. Zn was the regulated HM with the highest concentration in the soil (45.33 mg kg^{-1}), followed by Cu, Ni, Pb and Cr with the lowest concentrations associated to Cd and Hg. The statistical parameters show that over 50% of the data are below the mean with skewness higher for the lowest mean values, such as Hg, and Cd and always positive, which means that most of the rare cases are associated to high values. All kurtosis values are positive, which is indicative of a higher concentration of values around the mean when compared with the normal Gauss curve.

3.1.Soil analyses vs. regulations

All HM mean values were below the thresholds indicated by the Spanish Royal Decree 1310/90 and the most restrictive limits described by the EUDWD (European Union Draft Working Document (EC, 2000)). If each HM is independently evaluated, 90% of the samples were always below the thresholds indicated for acid soils by the Spanish Royal Decree 1310/1990. However, all the maximum values for the evaluated soils were above the thresholds with the exception of the Hg, when compared with the values provided by the Spanish Royal decree for acid soils. Only 151, 55, 19, 11, 9 and 2 samples of our 2597 total samples were over the limits for Ni, Cu, Zn, Pb, Cr and Cd stipulated by the Spanish regulations, which represents a percentage of around 5.8%, 2.1%, 0.7%, 0.4%, 0.3% and 0.1% of all evaluated samples, when each HM is independently taken into account, respectively. If the EUDWD draft limits are considered, then around 41.0% of the samples could not receive SS, due to their pH level being lower than 5, irrespective of the HM concentrations. From the 1066 samples with pH levels below 5, 473 samples meet the limits set by the EUDWD draft regarding pH (between 5 and 6). Out of the remaining 59.0% of samples with pH above 5 and if each HM is individually taken into account, the number of soils that could not receive SS was 28, 464, 352, 140, 123 and 393 for Cd, Ni, Zn, Hg, Cr, and Cu, respectively. This makes a total of 1.8%, 30.3%, 22.9%, 9.1%, 8.0% and 25.7% of the soil samples with respect to the samples with pH above 5 and a 42.1%, 58.9%, 54.6%, 46.4%, 45.8% and 56.2% of the total number of samples (including those samples with pH below 5) that will not be able to receive SS as fertiliser for each respective HM.

When all limits are considered together, around 93.3% of samples (2424) meet the 1310/90 Spanish regulation (based on the Spanish implementation of the currently approved EU regulation - Council Directive 86/278/EEC *of 12 June 1986 on the protection of the environment, and in particular of the soil, when SS is used in agriculture*) and therefore 6.7% (173 samples) did not fulfil the Spanish regulations. Around 42.7% of the soils that do not fulfil the current regulations (173 out of 245 determinations) do so for >1 HM and this makes it advisable to evaluate the relationship between the concentrations of different HMs, when trying to find an underlying factor that affects them. When the EUDWD is taken into account, only 28.7% of soils could receive SS. For each pH interval, the samples that could not receive SS were 41.0%, 63.0%, 2.6% and 5.3% for pH below five, 5–6, 6–7 and above 7, respectively. If each determination is evaluated, 1500 samples will not be able to receive SS, which means around 91% (1500/784) of the samples did not fulfil the EUDWD because of more than one HM, if only samples with pH above 5 are considered.

3.2. Characteristics of the study site

When a comparison of each HM between the soil groups that fulfil and do not fulfil the Spanish regulation RD 1310/90 was performed, it was found that they were all significantly different with the exception of Cd and Hg (Table 2). Differences between those soils that fulfil the Spanish regulation RD 1310/90 were: one and a half-fold for Pb, two- fold for Zn, 3-fold for Cr, 4-fold for Cu and 5-fold for Ni.

However, the Box-Whisker figures (not shown) of each HM reveal that there is an overlapping between those soils that fulfil and those that do not fulfil the Spanish regulation. Thus, it is not possible to discriminate between the soils that are able to receive SS under the current regulations, based on any particular HM. Therefore, a multivariate approach was performed. A factor analysis (with principal components extraction, followed by VARIMAX rotation) was used to investigate whether the HMs with significant differences (Ni, Pb, Zn, Cr and Cu) represent identifiable underlying factors in those samples with soil pH lower than 7. Factor analysis (Fig. 2a) showed that total soil concentrations of Cu, Ni and Zn are related and described by the factor 1 with Pb described by factor 2. This means that those samples with high levels of Zn also have high levels of Ni and Cu and they are independent of samples with high concentrations of Pb. Fig. 2b shows that those samples that do not fulfil the Spanish regulations have high values for the scores of factor 1 and/or factor 2.

On the other hand, there are 40.1% of samples that have more than one HM that does not fulfil the Spanish regulations to receive SS. Out of 173 soils that do not fulfil the Spanish regulations, there are 116, 40 and 17 samples that have one, two or three HMs that exceed the values stipulated by the Spanish regulations, respectively. Fig. 3 shows the results of a factorial analysis carried out for the concentrations of Cr, Pb, Ni, Cu and Zn within those samples that do not fulfil the RD 1310/90 regulation. The total soil concentrations of Ni, Cu and Zn are related to Factor 1, which means that those samples with high values of Ni also had high values of Cu and Zn. Factor 2 is positively and negatively highly correlated with the levels of Cr and Pb, respectively. This fact makes it advisable to carry out a hierarchical cluster analysis to draw the dendogram described in Fig. 4.

The cluster analysis allows us to divide those samples that did not fulfil the current regulations into groups. At the indicated similarity level of 12, a division of samples into four groups was chosen; the two smaller groups (group 1, group 2) consisting of 9 and 11 soil samples, respectively. The next largest group is formed by 130 soil samples and the remaining group is composed of 23 soil samples, being groups

3 and 4, respectively. The groups were based on the metal that exceeds the Spanish regulation limits and the number of HMs that the soil samples have above those limits.

Fig. 5 shows the scores reached by the soils that do not fulfil the Spanish regulations for both factors extracted as explained in Fig. 3. Fig. 4 and Table 3 show that groups 1 and 2 are those samples that have highest Cr and Pb values, respectively. Soil groups 3 and 4 are those that have levels of Ni, Zn or Cu higher than the specifications given by the Spanish Royal Decree; particularly most of the soil samples of Group 4, which have the highest levels of factor 1 because they exceed the levels for three HMs at the same time. Within group 3, the samples with the highest scores are those that have both Ni and Cu as the HMs that exceed the limits, the rest of the samples of this third group did not fulfil the SS regulations for one HM, mostly Ni.

A description of the location of those samples that exceed in 1, 2 and 3 metals in each county and the percentage of soil samples per county that do not fulfil the Spanish regulations can be seen in Fig. 6. As was previously indicated, Ni is the HM that limits the use of SS in the highest number of plots per county and for this reason Ni limited the use of SS in a higher number of counties than the rest of the HMs. Pb, Cu, Zn and Cr limits the use of SS based on RD 1310/90 in some specific counties, such as Santiago de Compostela or Moeche (NW of Galicia), which are also the areas that have plots that do not fulfil the requirements of RD 1310/90 in respect of two or three HMs.

4. Discussion

Soil HM ranges found in this study were within those generally described for Cd (Page et al., 1981) (0.01–3 mg kg^{-1}), Pb (Pais and Jones, 1997) (3–189 mg kg^{-1}), Cr (Alloway, 1995) (0.3–10,000 mg kg^{-1}) and Hg (Pais and Jones, 1997) (0.01–1.8 mg kg^{-1}). However, the essential elements for plants like Ni (Alloway, 1995) (1–100 mg kg^{-1}), Cu (Domínguez-Vivancos, 1997) (3–100 mg kg^{-1}) and Zn (Barber, 1995) (10–300 mg kg^{-1}) have a broader range than those values given in the literature for soils. The association of high levels of HMs with soils derived from ultrabasic and basic rocks soil parent material (Ross, 1994; Díez-Lázaro et al., 2002; Candeias et al., 2011) could explain the broader range of Ni and Zn as the factorial analyses carried out in our study demonstrate.

Our results showed that N 90% Galician soils are suitable to receive SS under the current regulations. However, the fact that all mean and median values of the agrarian soils of the present study were below the HM baseline levels of all types of Galician soils (Macías-Vázquez and Calvo de Anta, 2009), indicates that most of the Galician soils are suitable to receive SS fertilisation without a significant increase of the usual levels of HMs in soils for each type of rock parent material and if acceptable doses of SS with adequate quality (low HM levels) are applied in order to fulfil crop requirements. Recent revisions of the benefits of using high quality SS as fertiliser are provided by Diacono and Montemurro (2010), Lu et al. (2012) and Smith (2009).

Considering the mean chemical characteristics of the SS in Spain (Mosquera-Losada et al., 2010) stabilised by anaerobic digestion, we can estimate the amount of SS that should be applied in a soil for a given N rate (EPA, 1994). Once known the loading rate and the HM concentrations of SS, we can estimate the real inputs of HM per application and the minimum number of applications that SS could be used to reach the maximum concentrations in soil of HMs allowed by the Royal Decree 1310/90 in acid soils, assuming no relevant leaching or crop extractions of HMs from soils (McGrath, 1987). Anaerobic SS inputs of 200 kg of total N ha^{-1} (EPA, 1994) in a soil, implies a total increase of 1.26, 0.37, 0.05, 0.01, 0.16, 0.001 and 0.03 mg of Zn, Cu, Ni, Cd, Pb, Hg and Cr per kg^{-1} of soil and application, respectively (standard soil density of: 1.1

Mg m3 and a depth of 0.25 m (RD 1310/90)). These figures allow a minimum number of applications of 82, 89, 304, 94, 253, 491 and 2474 (usually years because SS is annually applied) for Zn, Cu, Ni, Cd, Pb, Hg and Cr in a mean soil, respectively. Therefore, the essential nutrients Zn, Ni and Cu are the most limiting for applying SS, as found McGrath (1987) in soils evaluated after long term SS inputs.

Among all soil parameters that could modify soil HM fates, only soil pH is currently included in most of the regulations for SS use in agriculture. Soil pH directly affects HM availability (Parat et al., 2005; Lu et al., 2012) and therefore their fate: mainly leaching or crop extraction. It is known that most HMs precipitate in neutral and basic soils (McGrath, 1987), and only soil movement (i.e. ploughing) could explain HM depletion (McGrath, 1987). On the contrary, in acid soils and soils with a gradual reduction of organic matter, an effect called "sludge time bomb" can appear. Sludge time bomb means that more soluble forms of HM can be released from soils as organic matter mineralization happens and, later on, taken by crops. This could explain, for example, why soil total bacterial populations are initially improved after nutrient supplies with SS inputs and later depleted when HMs availability is increased in soils (Guiller et al., 2009). The "sludge time bomb" concept, that could be more associated to acid soils, makes important to propose criteria for sludge inputs in soil ensuring that mean soil values of HMs for each type of soil are not surpassing. Having soil mean values for each type of parent rock material as a criterion for applying SS probably ensures that microbial and natural plant population can survive in those already existing environments, therefore promoting sustainability and preserving beta biodiversity among different types of soil conditions. Moreover, preserving soil mean values of HMs for each type of rock parent material, will also be adequate for sustainability of agricultural production of human food, as these soils were usually used for crop production.

The current EU and Spanish regulations are based on pH at a broad range. Most of the Galician soils have an acid pH, as happen with the agrarian soils evaluated in this study (only 1.5% of the soil samples have a pH above 7). The EUDWD draft does not mention the levels of HMs below which the sludge could be applied if pH is below 5, which directly affects 41% of the Galician soils. Even though, soil acid pH is a key factor increasing soil HM solubility (Parat et al., 2005) and therefore its potential to be leached and uptaken by crops, pH indicator has several concerns as a tool for SS applications for acid soils. Natural Galician soils are generally below pH 5 when no lime is applied, mainly due to the rainfall regime and crop extraction. Recommendations for applications of lime every four years in Galicia are usually based on the reduction of the percentage of saturation of Al to below 20%, which can be frequently obtained if the soil pH is around 5.5. If soil pH is artificially increased with lime applications, SS inputs are allowed and therefore the "sludge time bomb" process could be easily started, once liming is ceased. Of the soils analysed in the current study, 41 and 88% had a pH below 5 and 6, respectively. This means that if SS was used because soil pH is above 5 and no lime was applied for a long period of time, then soil pH could be easily reduced. On the other hand, even though most HMs are more available at low soil pH, some such as Cu could become less available below 5 than over this value (Porta, 2010). Therefore, it may be better to take into account the existing baseline levels of HMs in soils and to control the maximum dose permitted to be added as a percentage of that existing baseline levels. This will allow the application of more targeted amounts of SS, depending on the type of soil and the soil parent material, avoiding significant increases in soil HMs in a controlled way, which will protect the ecosystem at the same time, and probably reduce the negative long term impacts of SS inputs on soil total and nitrogen fixing microorganisms (Guiller et al., 2009). Baseline mean levels for each type of soil will probably allow the application of SS in >44.37% (473/1066) of soils with pH below 5, as all the mean HMs are below the lowest thresholds established by the EUDWD draft regulation. On the other hand, the reduction in sludge application advised by EUDWD has avoided the necessity to implement the EUDWD draft regulation. Moreover, it is also important to recommend other practices, such as the use of high quality sludge, the incorporation of SS in soils with ploughing to avoid a gradient of SS and to forbid direct grazing after applications, as animals consume large amounts of soil (ranging between 182 and 803 kg per year in dairy cows (Herlin and Andersson, 1996)) and therefore of HMs. The frequency of application on the same land should also be reduced.

5. Conclusions

Soil HM ranges found in this study were within those generally described for Cd, Pb, Cr and Hg, with the exception of Ni, Cu and Zn that have a broader range than those values given in the literature for soils. The high levels of Ni, Cu and Zn are explained by the fact that some soils derived from ultrabasic and basic rocks soil parent material. More than 90% of Galician soils are suitable to receive SS fertiliser under the current regulation RD 1310/90, but only 28.7% fulfil the EUDWD requirements. Most of the samples that do not fulfil the Spanish current regulation are associated to basic and ultrabasic rocks that define natural environments with specific plants and soil microorganisms already adapted to these levels of HMs. In order to apply more sustainable practices for agricultural production, it is proposed to take into account the mean HM levels of the soil for each HM trying not to surpass the mean levels of the soils derived from the different parent rock material, after considering human health risks. Moreover, this recommendation would respect the original environment of the soil that acts as a habitat for different organisms, preserving beta biodiversity.

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Table captions

Table 1 Statistical summary of selected properties for 2597 Galician soil samples used in this study compared with current and draft regulations. Min: minimum value, Max: maximum value; SD: standard deviation; Notice that there were 1558 samples (41%) out of 2597 with pH below 5. Values between brackets represent the percentage of all samples for each directive.

Property \ Value	Min-Max	Mean	SD	Median	Skewness	Kurtosis	90 percentile
pH-H_2O	3.44 – 10.22	5.21	0.72	5.17	1.25	5.67	6.05
Cd (mg kg^{-1})	0.01 – 2.50	0.05	0.15	0.01	5.63	50.07	0.10
Ni (mg kg^{-1})	0.001 – 169.50	12.20	14.28	8.90	2.31	10.80	27.70
Pb(mg kg^{-1})	0.01 – 118.50	10.21	12.13	6.60	1.75	5.34	27.12
Zn (mg kg^{-1})	0.01 – 306.70	45.33	30.46	41.20	1.58	6.39	81.92
Hg (mg kg^{-1})	0.01 – 0.80	0.054	0.05	0.05	5.33	52.08	0.10
Cr (mg kg^{-1})	0.01 – 236.94	10.03	16.54	5.80	4.87	37.09	22.32
Cu (mg kg^{-1})	0.01 – 212.00	16.38	17.98	12.30	3.73	23.98	34.70

	Legal requirements for use sewage sludge in soils. Samples below/above the limits														
	R.D. 1310/1990 *(Directive 91/271/EEC)*						3º Draft Working document on sludge (EU)								
	pH <= 7	n=2559		pH > 7	n=38		5<= pH <6	n=1230		6<= pH < 7	n=263		pH > 7	n=38	
	limit	below	over	limit	below	over	limit	below	over	limit	below	over	limit	below	over
Cd (mg kg^{-1})	*1*	2557 (98.4)	2 (0.1)	*3*	38 (1.5)	0 (0)	*0.5*	1203 (46.3)	27 (1.0)	*1*	262 (10.1)	1 (0)	*1.5*	38 (1.5)	0 (0)
Ni (mg kg^{-1})	*30*	2408 (92.7)	151 (5.8)	*112*	38 (1.5)	0 (0)	*15*	770 (29.6)	460 (17.7)	*50*	261 (10.1)	2 (0.1)	*70*	36 (1.4)	2 (0.1)
Pb(mg kg^{-1})	*50*	2548 (98.1)	11 (0.4)	*300*	38 (1.5)	0 (0)	*70*	1230 (47.4)	0 (0)	*70*	263 (10.1)	0 (0)	*100*	38 (1.5)	0 (0)
Zn (mg kg^{-1})	*150*	2540 (97.8)	19 (0.7)	*450*	38 (1.5)	0 (0)	*60*	880 (33.9)	350 (13.5)	*150*	262 (10.1)	1 (0)	*200*	37 (1.4)	1 (0)
Hg (mg kg^{-1})	*1*	2559 (98.5)	0 (0)	*1.5*	38 (1.5)	0 (0)	*0.1*	1090 (42.0)	140 (5.4)	*0.5*	263 (10.1)	0 (0)	*1*	38 (1.5)	0 (0)
Cr (mg kg^{-1})	*100*	2550 (98.1)	9 (0.3)	*150*	38 (1.5)	0 (0)	*30*	1111 (42.8)	119 (4.6)	*60*	263 (10.1)	4 (0.2)	*100*	38 (1.5)	0 (0)
Cu (mg kg^{-1})	*50*	2504 (96.4)	55 (2.1)	*210*	38 (1.5)	0 (0)	*20*	837 (32.2)	393 (15.1)	*50*	263 (10.1)	0 (0)	*100*	38 (1.5)	0 (0)

Table 2. t student comparison between samples that do, or do not fulfil the Spanish regulation RD 1310/90. ***: p<0.001.

	Samples that fulfil the Spanish Regulation RD 1310/90		Samples that not fulfil the Spanish Regulation R D 1310/90		
	Mean	EE	Mean	EE	t
Cd (mg kg^{-1})	0.05	0.003	0.05	0.02	-0.79
Ni (mg kg^{-1})	9.64	0.19	47.05	1.57	-23.70***
Pb(mg kg^{-1})	9.77	0.23	14.90	1.46	-3.46***
Zn (mg kg^{-1})	41.93	0.52	88.82	3.47	-13.36***
Hg (mg kg^{-1})	0.05	0.001	0.05	0.004	1.26
Cr (mg kg^{-1})	8.95	0.28	25.07	2.61	-6.14***
Cu (mg kg^{-1})	13.63	0.23	52.63	3.00	-12.95***

Table 3. Number of samples and heavy metals above limits in each group of unfulfilled metals soil samples.

	Metals above limits in samples into the group (Nº of samples in branches)			
Number of metals above limits	Group 1	Group 2	Group 3	Group 4
1	Cr (3)	Pb (9)	Cd (2) Ni (94) Cu (8)	
2	Cr, Ni (6)	Pb, Ni (2)	Ni, Cu (26)	Ni, Zn (2) Ni, Cu (4)
3				Ni, Zn, Cu (17)
Nº of samples in the group	9	11	130	23

Figure captions

Figure 1. Galician dominant land use (left) and number of soils sampled by county (right) in the current experiment

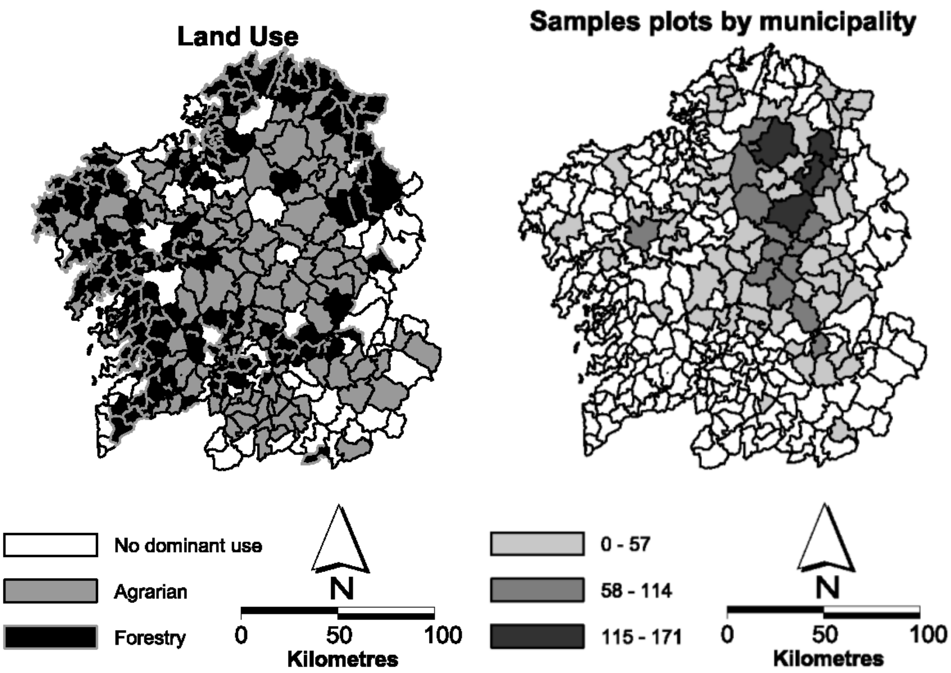
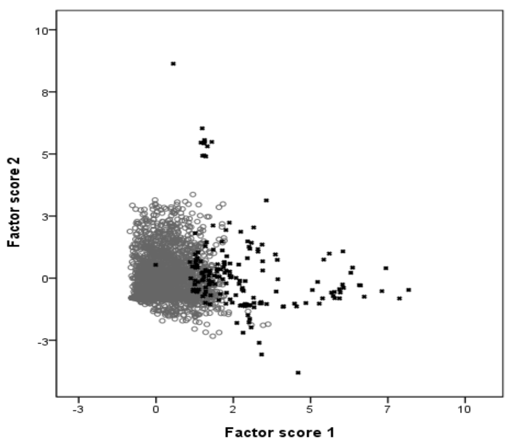
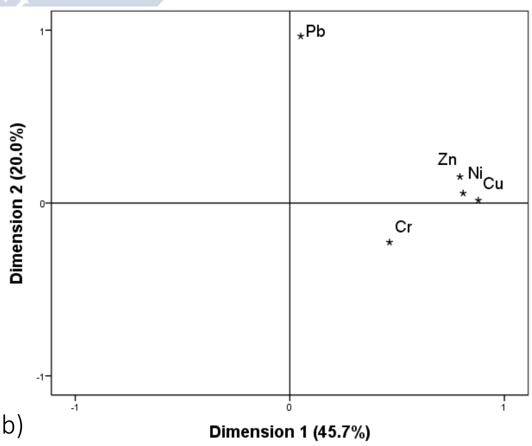


Figure 2. Factorial analysis of heavy metals of all studied samples (Figure 2a) and score factor plot of samples (Figure 2b). Dark spots represent those soils with limits above the Spanish regulation. Varimax rotation, KMO=0.69



a)



b)

Figure 3. Factor analysis of heavy metals with samples that do not fulfil the Spanish RD 1310/90. Varimax rotation KMO=0.56.

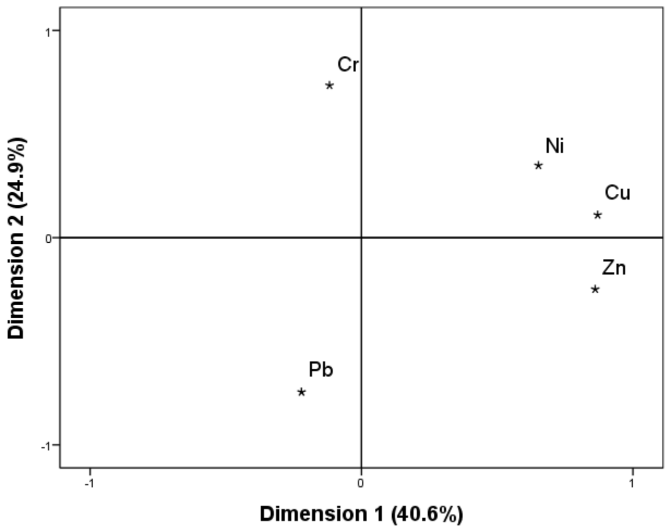


Figure 4. Dendogram performed from cluster analysis applied to the standardised data of those plots that do not fulfil the Spanish regulation.

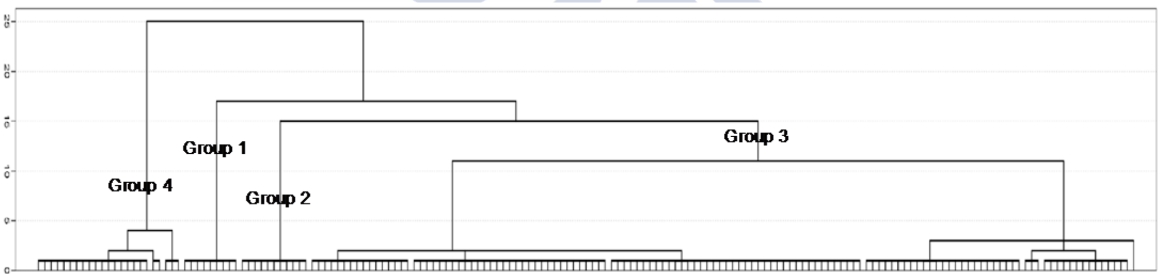


Figure 5. Score factor plot for analysis of soil samples that did not fulfil heavy metals. Overprinted numbers represent the number of metals that the sample does not fulfil.

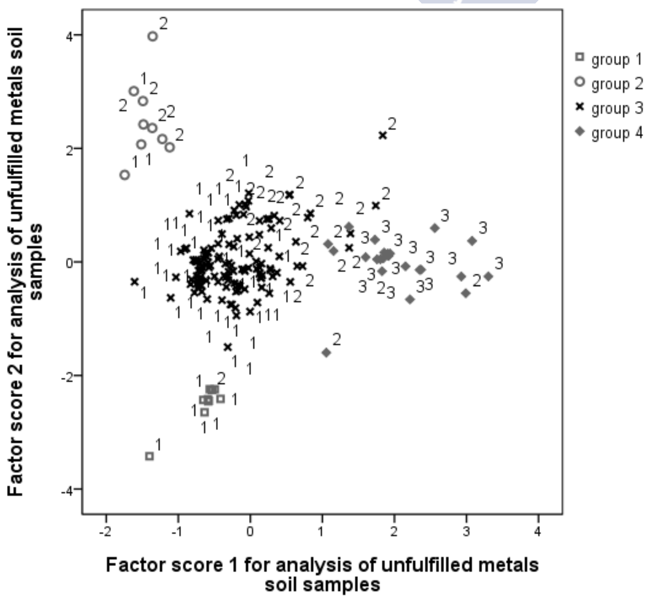
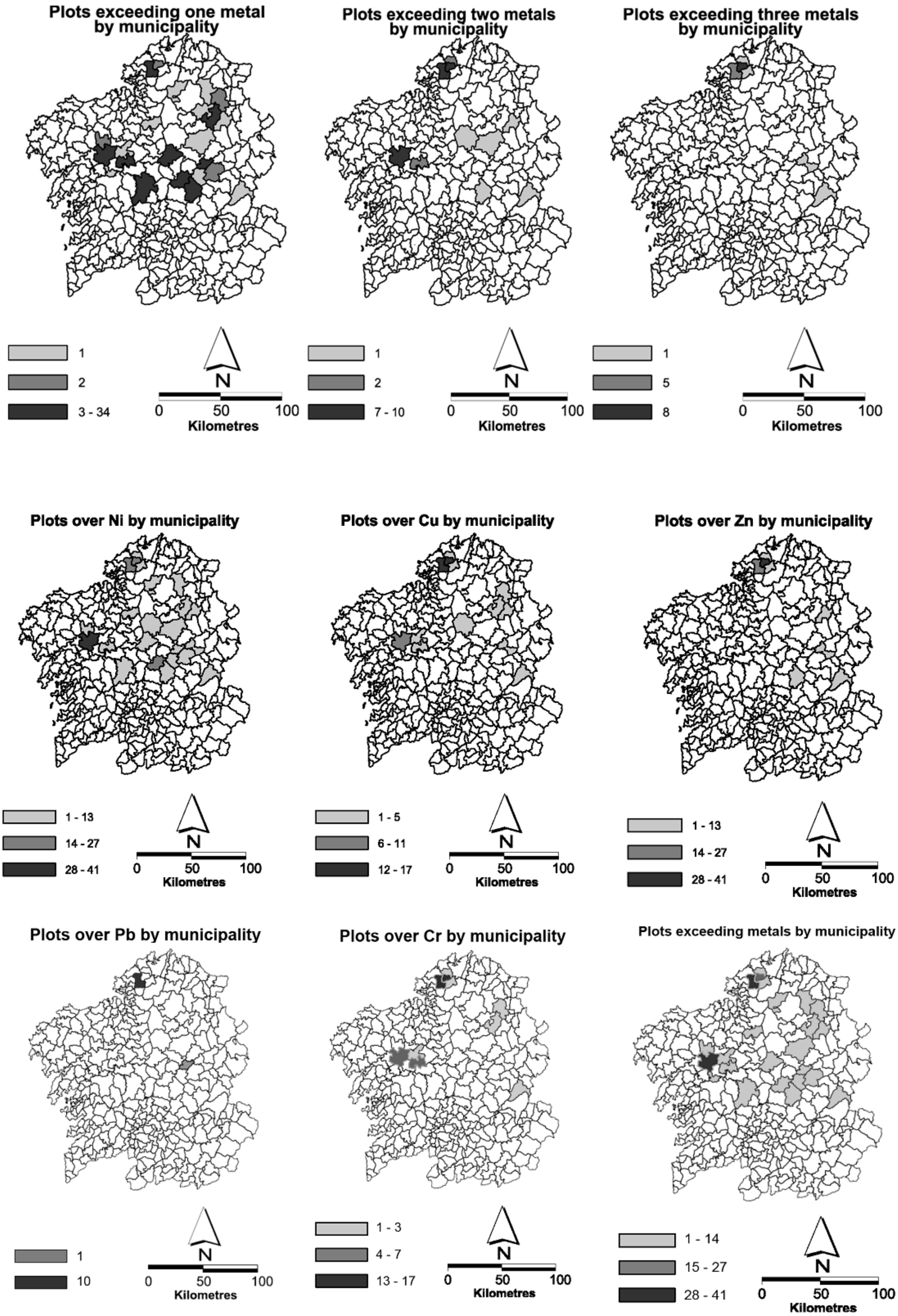


Figure 6. Counties with plots that exceed the limits established by the RD 1310/90 in one, two and three heavy metals in Galicia.



Capítulo 2. Proposing policy changes for sewage sludge applications based on zinc within a circular economy perspective

Abstract

Sewage sludge production has been increased in the last years as cleaning residual waste waters has been making compulsory in all small, medium and large size European villages and cities since 2005. Most adequate disposal of sewage sludge sludge should be agricultural soils to improve their fertility. However, the higher level of heavy metals in sludge than in soils makes obligatory to consider the inputs of heavy metals in soil as have been done by regulation 86/278/EEC and more recent documents (EU 3rd Draft Directive or the Environmental, economic and social impacts of these of sewage sludge on land (EC, 2008) proposes. Sewage sludge application is restricted by the mean amount of heavy metals applied with sewage sludge in soil in a 10-year period and the concentrations of heavy metals in the sludge and soils. This paper aims at evaluating the current regulation and the documents of the EU developed to improve the sewage sludge current regulation in the application of the regulated heavy metal with the higher sewage sludge concentration, the zinc after with different scenarios of the application of different doses and types of sewage sludge to fulfil crop needs. Main results highlighted the adequacy of using compost as amendment and anaerobic and pelletized sludge as fertilisers to reduce future possible soil contamination. Sewage sludge regulations should aim to reach mean values of Zn to ensure soil microbian health and sustainability adequate levels of Zn in crops or pasture. Low quality sewage sludge application should be reduced in frequency or doses, allowing soils to get benefits of increasing soil organic matter without strong changes in Zn soil contents.

1. Introduction

The implementation of the Urban Waste Water Treatment (EU, 1991) in all EU Member States has increased the quantities of sewage sludge requiring disposal. From an annual production of around 5.5 million Mg of dry matter (DM) in 1992, the European Union headed towards nearly 9.8 million Mg in 2012 (Eurostat, 2016). The fertilizer utilization is one of the most promoted uses of sewage sludge among scientists and policy makers in order enhance the recycling of sewage sludge nutrients (EU, 1991; Mosquera-Losada et al., 2011a; Mosquera-Losada et al., 2011b) because, if adequately managed, improves soil fertility increasing soil organic matter (BOE, 1990) therefore fulfilling bioeconomy and circular economy requirements. The stabilization process of sewage sludge modifies its fertilizer value and therefore the dose of application to fulfill crop needs (EPA, 1994; Mosquera-Losada et al 2010). The form of stabilization is usually selected depending on the volume of sewage sludge production and if composting is chosen it depends on the existence of materials or residues to mix within the area where the sewage sludge is produced (Mosquera-Losada et al., 2010). The most common and promoted treatments of sewage sludge to reduce microbian contamination and odors as well as to favor the agronomic use are anaerobic digestion and compost with is accompanied of a short and longer time of treatment, respectively. After anaerobic treatments, sewage sludge pelletization is useful when the volume reduction is pursued to decrease sewage sludge storage and transport costs, while improving soil application facilities.

The higher levels of heavy metals, pathogens and no-desirable organic compounds in the sludge than in the soil also causes environment concerns to the population, policy makers and scientists. Heavy metal limitations to use sewage sludge were established by the European Commission in 1986 (EU, 1986) and summarized by the Join Research Centre in 2012 (JRC, 2012). Nowadays, there are three conditions that limit the use of the sewage sludge in agriculture: (i) maximum concentration values in the sludge (ii) maximum concentration values in the soil and (iii) maximum amount of heavy metal that can be applied as a mean for a period of ten years in kg per hectare and year.

However, there is an agreement between policy makers, scientists and population that heavy metals thresholds should be further limited by modifying the EU Directive 86/278/CEE (EU, 1986). For this reason, the European Commission delivered a draft working document in 2000 (EC, 2000), that is still under discussion and that was not finally implemented due to the costs and the problems associated with the proposed more restrictive limits. The European Commission (EC, 2008) in the "Environmental, economic and social impacts of the use of sewage sludge on land. Final Report" document has evaluated these limits and posed several scenarios with less and more restrictive limits than the before mentioned draft (EC, 2000), estimating the implementing cost they have. In both drafts, sewage sludge application will be banned in soils with pH below 5.

Producing high quality sewage sludge is the first issue to be dealt with in order to use a residue in a sustainable form to improve soil fertility. Secondly, the evaluation of soils is highly relevant because the acidity and heavy metal baseline levels are essential to evaluate the fertility needs of the soil as well as the risk of sewage sludge inputs into the soil and the subsequently possibility of heavy metals reaching human beings causing health problems. The evaluation is more relevant in those regions with high demand of fertilizers due to their good environment growing conditions (temperature, humidity, radiation) and with acid soils due to the fact that heavy metals are more available in acid soils (Porta et al, 1999). In Europe, there is a large amount of soils that limits plant growth due to its acidity (Clea et al., 2014), where the dominance of natural factors determining the acid pH of agricultural soil at the continental scale (Salminen et al., 2005) should be compensated by the addition of calcareous amendments, that can be partially replaced by sewage sludge. Zn is the heavy metal with higher sewage sludge concentration from those regulated (EU, 1991; López-Díaz et al., 2007). But, at the same time, Zn is an essential trace metal for animals and plants. Zinc deficiency occurs usually associated to excessive calcium in soil linked to high soil pH (Tóth et al., 2016). Zn can be toxic in excess being animal exposure typically associated to dietary ingestion (Tóth et al., 2016).

Acidic Galicia soils could be considered representative of a large part of the Atlantic biogeographic region of Europe (EC, 2005; EEA, 2003) due to its acidity and the large agronomic and livestock production it has. This Spanish region which occupies around 3 million ha (IGE, 2011), producing 53% of wood in Spain (data from 2012)) (INE, 2015) but also produces 34% of the Spanish milk supply (data from 2013) mainly based on forage (crops like maize) and grasslands (INE, 2015). Currently, 97% of sewage sludge produced in the region (188.384 t in 2014) is valorized as fertilizer or soil improver. The aim of this study is to evaluate the current and foreseeable limits of sewage sludge application considering the current and future legislation of the most abundant heavy metal in the sewage sludge (Zn) considering the existing levels in acid soils in Galicia

2. Materials and Methods.

2.1. Characteristics of the study site

The study was carried out in Galicia, a Spanish region placed in the southwest part of the Atlantic biogeographic region of Europe.

2.2. Soil samplings and laboratory analyses

A set of 2597 soil samples from agricultural lands of Galicia (NW Spain), which were not previously fertilized with sewage sludge, were randomly taken at a depth of 25 cm as established 86/278/EEC regulation (EU, 1986). Out of this 2597, 2424 soil samples were selected for this study using as criteria the fulfilment of the 86/278/EEC regulation (EU, 1986) transposed in the Spanish regulation 1310/90 (BOE, 1990) for sewage sludge use in agriculture. Soil samples were mostly taken from the agrarian based counties of the Galician region, where more fertiliser is needed compared with the predominantly forested counties. The main agrarian activity in Galicia is related to forage crops and pasture to feed dairy and meat cows (IGE, 2011) and has a high rate of fertilizer needs and use. The number of hectares of the sampled plots represents approximately 1.5% and 20% of grasslands and agrarian soils (including forage crops and grasslands) of Galicia, respectively. Once taken, all soil samples were transported to the laboratory and later on air dried. Afterwards, soil samples were sieved through a 2 mm sieve. Water soil pH was measured (1:2.5) (Guitian and Carballas, 1976) and the concentration of Zn was analysed with the VARIAN 220FS spectrophotometer using atomic absorption (VARIAN, 1989), after a nitric acid digestion made in a CEM MDS-2000 microwave (CEM, 1994).

2.3. Statistical and Mathematical analysis

Descriptive statistics (mean, median, standard deviation, robust coefficient of variation, skewness, kurtosis, etc.) of the soil Zn were performed in the 2424 samples. The descriptive statistics include median absolute deviation (MAD), which is a robust method for evaluating dispersion. Statistical analyses were carried out with SPSS (PASW 18.0) for windows.

For the soil Zn study, soil samples were split in 4 groups of range of soil pH ($pH<=5$, $5<pH<=6$, $6<pH<=7$ and $pH>7$) because this allows to compare the samples within (i) the different limits for Zn given by 86/278/EEC (EU, 1986) regulation for soils, (ii) the European Union 3rd draft of the Directive (EUDWD) (EC, 2000), and those described in the Final Report "Environmental, economic and social impacts of the use of sewage sludge on land" (EC, 2008) in two scenarios, (iii) "Moderate changes"- (S1) (iv) "More significant changes" (S2) (Table 1). Even though the 3rd draft working document on sludge was not approved due to the lack of consensus between the different countries, we use the intervals of pH to compare it with the current soil values, because it could give us an idea of the next steps that will be taken in the future regulation of the use of sewage sludge in agriculture at European level. In case of soil without stablished limit (pH<=5) we compare the soil Zn concentration with those provided for soils with pH between 5 and 6.

Both regulations, 86/278/EEC (EU, 1986) and EUDWD (EC, 2000), indicate that a soil is suitable to receive sewage sludge depending on (i) sewage sludge Zn concentrations which has been already evaluated by Mosquera Losada et al. (2010) in Spain (Table 2), (ii) the amount of Zn added with sewage sludge (kg ha^{-1} year^{-1}) for a period of 10 years (Equation 1) and finally (iii) soil mean Zn concentration before adding sewage sludge.

$$\overline{\mathrm{ZnI}_{10}} = \sum_{\mathrm{i}=1}^{j} \mathrm{Zn_i} / 10$$

Equation (1): $\overline{\mathrm{ZnI}_{10}}$: Ten year average of Zn inputs (kg ha^{-1}) from year 1 to year j (kg ha^{-1} year^{-1}).

To evaluate the soil Zn evolution after sewage sludge applications, an estimation of the soil inputs of Zn was carried out with the mean values of Zn of the most common types of sewage sludge used in Spain: as anaerobic (AS), compost (CS) and pellets (PS) (Mosquera-Losada et al., 2010) and the sewage sludge doses needed to fulfil crop and pasture required with different degree of intensification. Sewage sludge Zn inputs (ZnI) for a given period were calculated based on Zn concentrations and the dose of sewage sludge needed to fulfill the most common nitrogen doses of 50, 100, 150 or 200 kg available-N ha^{-1}, thereafter mentioned as 50N, 100N, 150N and 200N, usually carried out in Atlantic region of Europe to fulfill crop requirements. Sewage sludge dose to fulfill N crop requirements was estimated based on the sewage sludge mineralization rates, and therefore N availability given by EPA (1994), where approximately only of 20% of total N is available if sewages sludge is anaerobic processed and pelletized, and 10% if composted sewage sludge is applied to the soil. ZnI was calculated for a period of 10 years as indicates the 86/278/EEC (EU, 1986) and EUDWD (EC, 2000) regulations as an average (Equation 1) or as a cumulative application in consecutive years (Equation 2)

$$ZnI_j = \sum_{i=1}^{j} Zn_i$$

Equation 2: ZnIj: Zn inputs (kg ha^{-1}) from year 1 to year 10; Zni:Zn input (kg ha^{-1} $year^{-1}$).

These estimations were also used to calculate the number of needed years (N) for a soil to achieve each current 86/278/EEC (EU, 1986) or proposed EUDWD (EC, 2002), S1 and S2 (EC, 2008) regulations (LV (limit given by regulations (kg ha^{-1} $year^{-1}$) (86/278/EEC, EUDWD, S1 or S2), considering the soil current levels (sZn) and the inputs carried out considering one application per year (Zn_i) as describes Equation 3.

$$N = [LV\text{-}sZn_s]/Zn_i$$

Equation 3. Estimation of the number of years (N) needed to achieve the Zn limit indicated by 86/278/EEC (EU, 1986) or EUDWD (EC, 2002) or S1 or S2 (EC, 2008). LV*:* Limit given by regulations (kg ha^{-1} $year^{-1}$) (86/278/EEC, EUDWD, S1 or S2); sZn_s *:* Mean Zn content in soil (kg ha^{-1} $year^{-1}$); Zn_i:Zn input (kg ha^{-1} $year^{-1}$).

The year to year Zn soil balance evolution of soil Zn (sZn) was estimated by summing up the total annual inputs of Zn and the already existing mean levels of Zn in the soil for each range of soil pH obtained in this study which was substracted by the estimated amount of extracted Zn carried out by the crop both expressed in kg per ha. Soil mean values for Zn were expressed as kg per hectare considering the initial mean concentration of Zn in soil (mg kg^{-1} obtained from the 2424 samples of the present study), an average soil bulk density of 1.12 Mg m^{-3} and a depth of 0.25 m (indicated by the Council Directive 86/278/EEC, (EU, 1986)) (Equation 4). Extracted Zn (eZn) was also considered based on a mean grass production in Galicia of 6000 kg ha^{-1} (Mosquera-Losada et al, 1999) and a mean baseline value of Zn in crops growing in no fertilized soils (16 mg Zn per kg of pasture (Mosquera-Losada et al., 2009). We estimated that extracted Zn by pasture in Galicia is 96 g ha^{-1} $year^{-1}$ (i.e. 16 mg Zn kg^{-1} x 6000 kg ha^{-1})

$$sZn_j = sZn_{j-1} + \sum_{i=1}^{j}(Zn_i - eZn_i)$$

Equation 4. Estimation of the balance of Zn soil content (SZn; kg ha^{-1}) from N=0 until year j; sZn_s : Mean Zn content in soil obtained in this study (kg ha^{-1}); Zn_i: Zn input (kg ha^{-1} $year^{-1}$). eZn_i: crop Zn extraction (kg ha^{-1} $year^{-1}$).

3. Results

Table 3 shows the results of the descriptive statistics of soil pH and Zn obtained from the 2424 samples selected in this experiment based on fulfilment of 86/278/EEC (EU, 1986). Water soil pH was acid (5.22 and 5.17 for mean and median, respectively), with a large range from 3.4 to 10.2. All kurtosis figures were positive, which is indicative of a higher concentration of values around the mean when compared with the normal Gauss curve. The mean concentration of Zn in the soil was 42.23 kg ha^{-1}, with a large range from 0.01 to 248.31. CVR (robust coefficient of variation) was high, which implies a high range of the mean and makes more interesting to represent the median and mean together. Median absolute deviation (MAD) is a measurement of the variability not influenced by the high values and also indicated that most of the rare cases are associated to high values.

Table 4 shows the results of the descriptive statistics of Zn obtained for each range of soil pH (pH<=5, 5<pH<=6, 6<pH<=7 and pH>7) from the 2424 soil samples considered in this experiment. Most of the samples that fulfill 86/278/EEC (EU, 1986) were acid (2138 samples involving 98% of soils). Therefore they should be compared with the acid soil limit values established by the Directive 86/278/EEC (EU, 1986). Moreover, around 88% of samples had a pH below 6. Zn mean soil was above the median, which is indicative that over 50% of the soils have Zn concentrations above the mean for Zn. All soils have different concentrations of Zn depending on soil pH. Soils with pH above 7 tend to have higher concentration of Zn than those soils with pH below 7. Zn concentrations in soils with pH below 5 were lower than those with a soil pH above. Soils with a pH between 6 and 7 fulfill EUDWD (EC, 2000) requirements for Zn. However, around 24.9% of soils of the group of pH between 5 and 6 were above EUDWD limits for Zn. Furthermore, if soils with a pH below 5 are taken into account and compared with the limit heavy metal values of soil pH between 5 and 6, we found that 21.3% of soil samples did not fulfill EUDWD (EC, 2000) requirements for Zn. Soils with a pH between 6 and 7 fulfill S1 requirements (EC, 2008) for Zn. However, around 2.3%, of soil samples for group of soil pH between 5 and 6 were above S1 limits for Zn. Moreover, if soils with a pH below 5 are taken into account and compared with the limit S1 values of pH between 5 and 6, we found that 1.9% of soil samples did not fulfill S1 requirements for Zn (EC, 2008). Regarding S2 (EC, 2008), we found that around 84,3%, of soil samples for group of soil pH between 6 and 7 were above S2 limits for Zn. Moreover, up to 69.7% of soils with a soil pH below 5 and 79.8% of those soils with a pH between 5 and 6 did not fulfill S2 requirements for Zn when the limit values for pH between 5 and 6 were considered. Only around 2,6% of soils with a soil pH above 7 not fulfill EUDWD (EC, 2000), S1 and S2 requirements.

Table 5 shows amounts of Zn which will be added annually to soil, based on a ten year average (kg ha^{-1} $year^{-1}$) according to different available-N inputs (50N, 100N, 150N and 200N) and type of sewage sludge (CS, AS and PS). As expected, high doses of CS (150N and 200N) caused that Zn reached the threshold provided by 86/278/EEC (EU, 1986) regulation earlier than the other sludge treatments and doses. Only low dose (50N) of AS based on a ten year average will not reach the limit value established in EUDWD (EC, 2000). The studied doses of CS, PS and AS, based on a ten year average will cause that soil Zn reaches the limit values established in EUDWD (EC, 2000), except 50N dose of AS if a continuous application of sewage sludge is carried out. When the current soil Zn concentrations are compared with the sum of Zn added by the different sewage sludge types and doses for a period of 10 years, we can see that Zn inputs were higher than soil concentration and therefore the main driven force regulating soil Zn content (Tables 3 and 5).

Figure 1 shows the diachronic evolution of Zn content in soil (sZn) within three pH intervals (<5, 5-6 and 6-7), considering different N doses and Zn extractions and types of sewage sludge. Crop extraction (eZn) is negligible when compared with the already existing soil Zn concentrations and Zn inputs in the first year. Zn extractions were lower than 0.02 % than those applied with sewage sludge. Zn mean in soil after 10 years of AS applications are below thresholds in soil for the Zn limit established in 86/278/EEC (EU, 1986) regulation. Moreover, Zn in soil after 10 years of application was close to the limit value established in EUDWD (EC, 2000) if 50N dose was added to the soil as AS. AS doses of 200N caused a surpass of the thresholds in soil for Zn established in EUDWD (EC, 2000) after four years of application in soil pH below 5, and after three years of application in soils with pH between 5 and 7. In the case of AS, doses of 200N caused a surpass of the soil thresholds for Zn established by S1 (EC,

2008) after ten, nine and two years of application in soils with a pH below 5, between 5 and 6, and between 6 and 7, respectively. CS applications of a dose of 50N for a ten-year period did not caused that soils reach the thresholds provided by the 86/278/EEC (EU, 1986) regulation and in the S1 (EC, 2008). However this threshold was surpassed after seven years when additions of 150N of CS were applied in all soil pH range. On the contrary, all evaluated doses of CS application caused that Zn in soil exceeded the limit value established in EUDWD (EC, 2000) draft for all range of soil pH evaluated. However, Zn concentration in soil applied with 100N, 150N and 200N doses of CS inputs for a period of 10 years exceeded the limit values established in S1 for all range of pH in soil. Finally, when a ten-year PS inputs were analyzed, we found that Zn concentration in soil was always below the limit values established by the 86/278/EEC (EU, 1986) Regulation in 50N, 100N and 150N as PS were applied. However, Zn in soil exceeded the limit values established in EUDWD (EC, 2000) after three years of continuous application of all doses of PS for a period of 10 years. Moreover, Zn in soil did not exceeded the limit values established in S1 (EC, 2008) when doses below 100N dose of PS were added to the soil for a period of 10 years. AS, CS, and PS applications were not allowed in S2 (EC, 2008), because initial Zn concentration in soil surpass limit value established.

Table 6 specifies the number of needed years to achieve soil Zn concentration heavy limit values provided by all groups of acid soil pH, considering the four N doses, the initial soil mean of Zn of each soil pH interval obtained in this study and the different types of sewage sludge. Thresholds (86/278/EEC (EU, 1986)) for Zn in soil were reached before 30 years with CS for all doses. A similar response was found in PS for this element with the exception of the smaller dose (50N). High doses of sewage sludge applied as PS (200N) or as CS (150N and 200N) would imply that soils reached the maximum levels of Zn in soils given by 86/278/EEC (EU, 1986) in a period below 10 years of sewage sludge application. However, periods between 11-20 and 21-30 years are needed if 200N or 150N doses of AS sewage sludge to surpass Zn limits provided by the 86/278/EEC (EU, 1986), respectively. Thresholds EUDWD (EC, 2000) for Zn in soil were reached before 20 years with CS for all doses. A similar response was found in PS and AS; EUDWD (EC, 2000) thresholds for Zn in soil were reached before 40 and 65 years, respectively. High doses of sewage sludge applied as PS (150N-200N) or CS would imply that soils reached the maximum levels of Zn in soil given by EUDWD (EC, 2000) in a period of sewage sludge application of less than 5 years if pH below 6 is considered. However, a period of 16 years were needed if high dose of sewage sludge (200N) are applied as anaerobic (AS) to surpass Zn limits provided supplied by the EUDWD (EC, 2000) in soils with pH above 6. S1 threshold was reached before 10 years of CS and PS applications with high doses (100N-200N) in soils with pH below 6 as well as with 200N of AS for the same pH interval. Similar response was found if S1 was considered (EC, 2008) with regard to the 6-7 pH interval. In the case of S2 (EC, 2008), the initial mean concentration of Zn in soil in all groups of soil pH linked to acid soils surpassed the limit for Zn established.

4. Discussion

Sewage sludge quality understood as the concentration of heavy metal it has varies between types of sludge and modifies the potential use of sewage sludge in agriculture (Mosquera-Losada et al., 2010). Even though, sewage sludge quality used in this study was relatively adequate compared with others (Mosquera-Losada et al., 2010) serious restrictions for its use as fertilizer appears if considering other sludge aspects regulated dealing with the maximum amount of Zn that can be applied into the soil as a mean during a ten year period and the concentration of Zn in the soil. Current regulation generally allows applications in soils during ten year period of PS and AS. However, the inputs of Zn with CS are seriously limited by the 10-year rule of the current regulation as consequence of the large amounts of compost needed to make available enough nitrogen to fulfil crop needs due to the low N concentration it has (Yongjie and Yangsheng, 2005). Other authors as Mosquera-Losada et al. (2010) found a similar result with evaluating the sewage sludge quality, which is justified by the lowest degree of mineralizable N when CS is applied compared with AS or PS (Mosquera-Losada et al., 2017). This makes advisable to use compost as a soil organic amendment (instead of a fertilizer) as the organic matter persistence in the soil will be longer than with AS and PS due to the high proportion of lignin in coming from the rest of the branches used to make compost with the sludge (Mosquera-Losada et al., 2017). Only low doses of N application with AS during 10 consecutive years will be allowed following the EUDWD regulation (EC, 2000), which is therefore more restrictive than the current regulation and seriously limits the use of any type of sewage sludge as fertilizer. Two solutions can be envisaged for the commitment of the 10 year and EUDWD rules (EC, 2000) one is to improve the sewage sludge quality and the second would be to apply sewage sludge with less frequency than a year and combining it with the application of inorganic fertilizers in the intermediate years to fulfil the need of N of the crops. An annual combination of inorganic N with that coming from sewage sludge could also be possible and will increase the availability of the sludge N thanks to the reduction of C/N when both types of fertilizers are combined together.

All soils samples fulfil current regulation EU Directive 86/278/EEC (EU, 1986) and Spanish RD 1310/1900 (BOE, 1991) (Zn: 150-450 mg kg^{-1}). However, all soils with a pH below 5 would not be able to receive sewage sludge following the EUDWD draft (EC, 2000) and only the 21.3% of soils can be considered adequate to receive sewage sludge if the limit between 5 and 6 is considered for those soils with a pH below 5 for EUDWD (EC, 2000). When the less (S1) and more restrictive (S2) JRC scenarios proposal (EC, 2008) are considered, only 1,9% but the 69.7% would not be allowed to receive sewage sludge when soil pH is below 5 if the limit values for pH between 5 and 6 are considered, respectively. Regarding Zn limits for soil pH between 5 and 6 a similar result was obtained (24.9, 2.3 and 79.8% for EUDWD (100 kg^{-1}), S1 (100 mg kg^{-1}) and S2 (20 mg kg^{-1}) would not be able to receive sewage sludge, respectively). Median Zn soil concentration found in this study (pH<5, 39.68 mg kg^{-1}; 5≤pH<6, 44.00 mg kg^{-1}) were really below the worldwide natural soils (Barber (1995): 10-300 mg kg^{-1}) and Kabata-Pendias and Pendias (2001): 10-105mg kg^{-1})), in the agricultural and grassland soils with pH below 7 in Spain (58.42 mg kg^{-1}) (Rodríguez Martín et al., 2009) and in the 98% of Galician (NW Spain) natural soils (Macías-Vázquez and Calvo et al., 2009 :100 mg kg^{-1}). Current agrarian Galician soils are cropped with Zn levels above S2 limit proposal (Mosquera-Losada et al., 2017), showing in several cases a pasture deficit on this heavy metal, which is needed and considered as micronutrient to feed animals. This deficit can be explained by the large amount of soil organic matter (SOM) in Galician soils (reaching values of 20%) as mentioned Sánchez-Rodríguez et al. (2002), as SOM adsorbs Zn (Alloway 1995; Kabata-Pendias and Pendias, 2001; McBride et al. 2004) and makes it not available to crops (Mosquera-Losada et al. 2009). High SOM content in Galicia can be explained by the high acidity of the soils which prevents from SOM mineralization due to the low soil bacteria presence to mineralize SOM. Considering SOM as part of the EU future regulations of sewage sludge in Europe may be relevant as 35.6% of grasslands and 25.8% of European soils are acidic (Clea et al., 2014).

The evaluation of the rules of the current and possible future regulations regarding the soil amount of Zn after different types of sewage sludge application showed that CS was also the most restrictive type of sewage sludge application when soil Zn content is considered after PS and AS were applied, with and without Zn crop extraction. For ten years or more years of different doses of sewage sludge applications in current regulation, CS (6-21 years) achieves the limit of Zn in soil by current regulation before than PS (10-41 years) and AS (16-68 years), respectively. In case of EUDWD (EC, 2000) for acid soils (pH<6), CS could be used for 4 years, PS for 8 years and AS for 13 year. CS was the worst sewage sludge treatment to meet the crop requirements as a fertilizer (Mosquera-Losada et al., 2017), increasing of Zn applied in soil, so that will be used with low application rate (Yongjie and Yangsheng 2005) as organic amendment to improve soil structure. Therefore EUDWD (EC, 2000) restricts the reuse of sewage sludge in agriculture as fertilizer more than Directive 86/278/EEC (EU, 1986) and in spite of seeking to encourage and fostering sewage sludge use as part of the circular economy context (EC, 2015). However, S1 permits sewage sludge use for more years than EUDWD in acid soils (pH<6). For S2, we could not use sewage sludge in agriculture, in spite of the high quality of Galician soils and the low sewage mean content of Zn found in the Spanish sewage sludge compared with other areas.

On the other hand, sewage sludge use have shown a clear benefit for non-food crops (forest trees) and grasslands when applied in the first or even in the three-four consecutive years (Mosquera-Losada et al., 2009, 2010, 2011a, 2012b, 2017) in long term experiments carried out in acid soils. Soil Zn contents were not dangerously increased even if larger amounts of N were applied (120N). Moreover, there are natural soils in Galicia and other European areas with high levels of Zn which are currently used for food production directly consumed by human beings. These soils have specific soil microbian populations that should be preserved. Therefore a good proposal to regulate Zn or any heavy metal inputs from sewage sludge in soils would be not to exceed the mean levels of the soils associated to specific parent materials (Mosquera-Losada et al. 2017), as well as making applications as amendments in non consecutive years which could help to enrich SOM and enhance benefits that this practice provides as soil fertility improvement, to maintain natural soil levels of Zn, and to fulfill animal requirements.

5. Conclusions

Based on the scenarios of the paper, the composted sewage sludge should be used as organic matter soil amendment, while those sewage anaerobic and pelletized sludges should be used as fertilisers to reduce future possible soil contamination. Sewage sludge regulations based on Zn heavy metal concentrations should aim to reach the mean values of Zn in soil usually associated to different rock parent material to ensure soil microbian health and sustainability adequate levels of Zn in crops or pasture. If sewage sludge quality is not improved, sewage sludge application should be reduced in frequency or doses, allowing soils to get benefits of increasing soil organic matter without strong changes in Zn soil contents.

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Table captions

Table 1. Limits of Zn concentration in soil (mg kg^{-1}) for each group of soil pH, established by the 86/278/EEC regulation (EU, 1986), the EUDWD (EC, 2000) and the Final Report (EC, 2008) in the both options: Moderate changes (S1) and More significant changes (S2).

	Zn (mg kg^{-1})			
Soil pH	86/278/EEC	EUDWD	S1	S2
pH<=5		-	-	-
5<pH<=6	150	60	100	20
6<pH<=7		150	150	20
pH>7	450	200	200	200

Table 2. Dry matter (% DM), total nitrogen (% N), and Zn (mg kg^{-1}) concentrations the different types of sludge studied (anaerobic, compost and pellet) obtained for a national survey carried out during two years in 45 sewage sludge plants in Spain by Mosquera Losada et al. (2010).

	Anaerobic (AS)	Compost (PS)	Pellet (PS)
% DM	19.81	75.78	92.91
% N	4.41	2.06	3.57
Zn (mg kg^{-1})	805.85	618.02	1086.64

Table 3. Statistical summary of selected properties for 2424 Galician soil samples that fulfil 86/278/EEC (EU, 1986) regulation, the main descriptive statistics of water pH and Zn in all evaluated soils. Min: Minimum value. Max: Maximum values. SD: Standard deviation. Med: Median. MAD: median absolute deviation. CVR: robust coefficient of variation. Skw: Skewness. K: Kurtosis.

	Min	Max	Mean	SD	Med	MAD	CVR	Skw	K
Soil pH	3.44	10.22	5.22	0.74	5.17	0.71	14,18	1.24	5.47
Zn (mg kg^{-1})	0.01	248.31	42.23	26.49	38.72	27.75	62,74	0.93	2.06

Table 4. Statistical summary of selected properties for 2424 Galician soil samples the main descriptive statistics of Zn (mg kg^{-1}) and pH in all evaluated soils for each group of soil pH and compared with current and draft regulations. For Zn means followed by a difference letter are significantly different at $P \leq 0.05$. Min: Minimum value. Max: Maximum values. SD: Standard deviation. Med: Median. MAD: median absolute deviation. Not EUDWD: number of soil samples that not fulfil EUDWD limit for Zn (EC, 2000). Not S1: number of soil samples that not fulfil S1 limit for Zn (EC, 2008). Not S2: number of soil samples that not fulfil S2 limit for Zn (EC, 2008). n: total number of soil samples that fulfils the current regulation in the different soil pH.

	Soil pH	Min	Max	Mean	SD	Med	MAD	Not EUDWD	Not S1	Not S2	n
pH	pH<=5	3.44	5.00	4.56	0.31	4.61	0.36	-	-	-	-
	5<pH<=6	5.01	6.00	5.46	0.28	5.44	0.34	-	-	-	-
	6<pH<=7	6.01	6.96	6.28	0.22	6.21	0.21	-	-	-	-
	pH>7	7.02	10.22	8.03	1.08	7.61	0.74	-	-	-	-
Zn	pH<=5	0.01	141.30	38.68 b	26.01	34.04	28.11	213	19	698	1001
	5<pH<=6	0.01	144.40	44.00 a	24.51	42.30	25.95	283	26	907	1137
	6<pH<=7	0.04	131.20	45.55 a	27.98	42.60	28.84	0	0	209	248
	pH>7	5.60	248.31	61.09ab	55.87	35.10	17.12	1	1	1	38

Table 5. Amounts of Zn which will be added annually to soil, based on a ten year average ($\overline{\mathrm{ZnI}}_{10}$) (kg^{-1} ha^{-1} year) according nitrogen dose (50N, 100N, 150N and 200 N, N=kg available N ha^{-1}) for compost (CS), anaerobic (AS) and pellet (PS) of sewage sludge. >EUDWD: Light grey colour indicate when amounts of Zn which are added annually to soil according nitrogen dose for compost, anaerobic and pellet of sewage sludge, based on a ten year average (kg^{-1} ha^{-1} year), exceed limit values for amounts of Zn (7.5 kg^{-1} ha^{-1} year^{-1}) established in EUDWD (EC, 2000). >86/278/EEC: Dark grey colour indicates exceed limit values for amounts of Zn (30 kg^{-1} ha^{-1} year^{-1}) established in Directive 86/278/EEC (EU, 1986).

	Anaerobic (AS)				Compost (CS)				Pellet (PS)			
Year	50 N	100 N	150 N	200 N	50 N	100 N	150 N	200 N	50 N	100 N	150 N	200 N
1	0,5	0,9	1,4	1,8	1,5	3,0	4,5	6,0	0,8	1,5	2,3	3,0
2	0,9	1,8	2,7	3,7	3,0	6,0	9,0	12,0	1,5	3,0	4,6	6,1
3	1,4	2,7	4,1	5,5	4,5	9,0	13,5	18,0	2,3	4,6	6,8	9,1
4	1,8	3,7	5,5	7,3	6,0	12,0	18,0	24,0	3,0	6,1	9,1	12,2
5	2,3	4,6	6,9	9,1	7,5	15,0	22,5	30,0	3,8	7,6	11,4	15,2
6	2,7	5,5	8,2	11,0	9,0	18,0	27,0	36,0	4,6	9,1	13,7	18,3
7	3,2	6,4	9,6	12,8	10,5	21,0	31,5	42,0	5,3	10,7	16,0	21,3
8	3,7	7,3	11,0	14,6	12,0	24,0	36,0	48,0	6,1	12,2	18,3	24,4
9	4,1	8,2	12,3	16,4	13,5	27,0	40,5	54,0	6,8	13,7	20,5	27,4
10	4,6	9,1	13,7	18,3	15,0	30,0	45,0	60,0	7,6	15,2	22,8	30,4

> 86/278/EEC   > EUDWD

Table 6. Number of needed years (N) for a mean soil to achieve Zn limit values of the 86/278/EEC regulation (EU, 1986), EUDWD (EC, 2000), S1 and S2 (EC, 2008) for a given nitrogen dose per year provided by a mean type of Spanish compost (CS), anaerobic (AS) and pellet (PS) of sewage sludge and added initial concentration mean of Zn in soil in group of soil pH (acid soils). Colors indicate year's interval to reach Regulation limits. "-x-", sewage sludge application not allowed because initial concentration mean of Zn in soil exceed the limit value established.

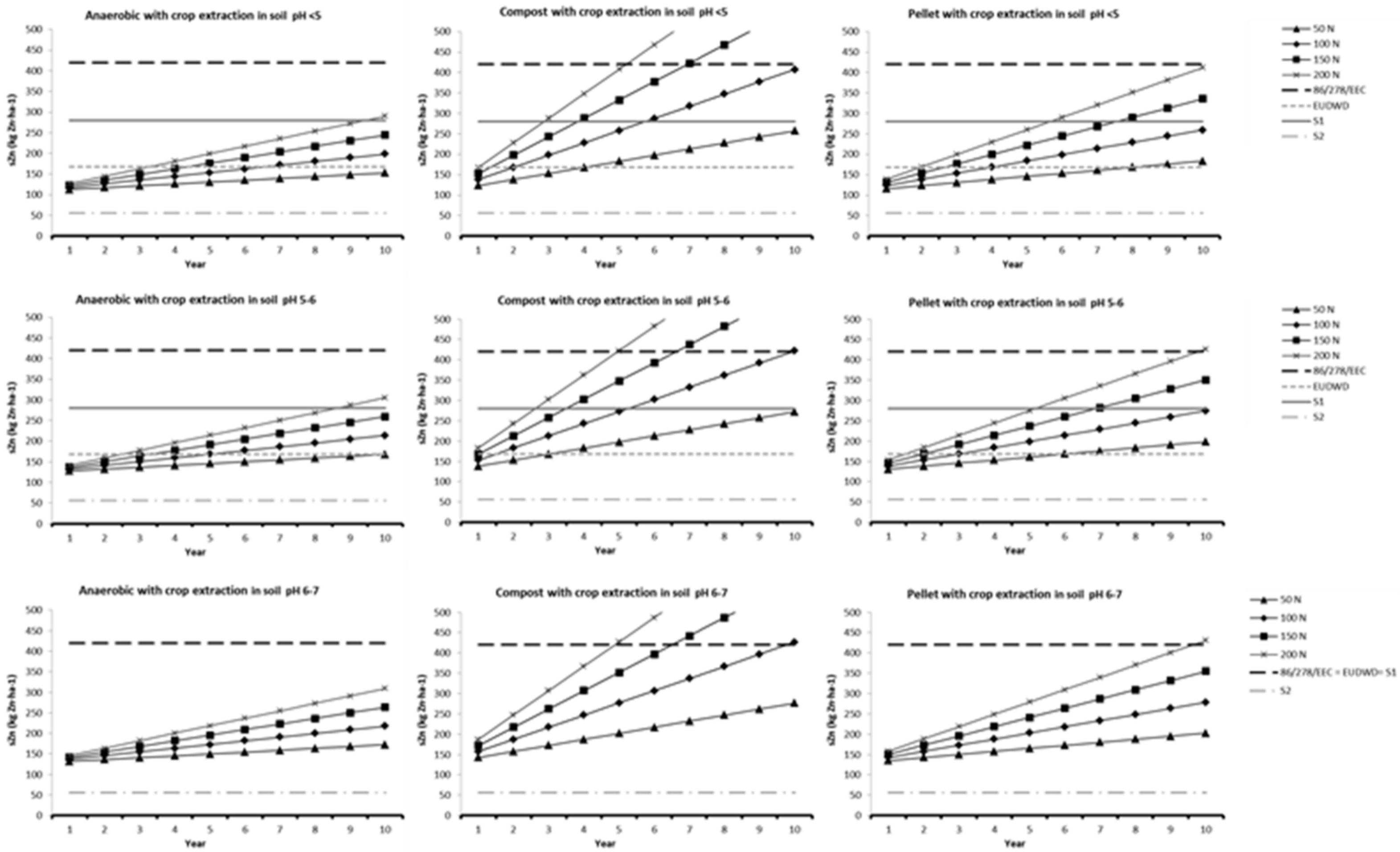
			86/278/EEC	EUDWD	S1	S2
Sludge	Soil pH	Dose	Zn	Zn	Zn	Zn
Anaerobic (AS)	pH<5	50N	68	13	38	-x-
		100N	34	7	19	-x-
		150N	23	4	13	-x-
		200N	17	3	9	-x-
	5<pH<6	50N	65	10	34	-x-
		100N	32	5	17	-x-
		150N	22	3	11	-x-
		200N	16	2	9	-x-
	6<pH<7	50N	64	64	64	-x-
		100N	32	32	32	-x-
		150N	21	21	21	-x-
		200N	16	16	16	-x-
Compost (CS)	pH<5	50N	21	4	11	-x-
		100N	13	3	7	-x-
		150N	9	2	5	-x-
		200N	7	1	4	-x-
	5<pH<6	50N	20	3	10	-x-
		100N	12	2	7	-x-
		150N	8	1	4	-x-
		200N	6	1	3	-x-
	6<pH<7	50N	19	19	19	-x-
		100N	12	12	12	-x-
		150N	8	8	8	-x-
		200N	6	6	6	-x-
Pellet (PS)	pH<5	50N	41	8	23	-x-
		100N	20	4	11	-x-
		150N	14	3	8	-x-
		200N	10	2	6	-x-
	5<pH<6	50N	39	6	21	-x-
		100N	20	3	10	-x-
		150N	13	2	7	-x-
		200N	10	1	5	-x-
	6<pH<7	50N	38	38	38	-x-
		100N	19	19	19	-x-
		150N	13	13	13	-x-
		200N	10	10	10	-x-

0-10 years	11-20 years	21-30 years	>30 years	-x-

.

Figure captions

Figure 1. Graphics show evolution (sZn) of initial content for Zn in soil (acid soil) according Nitrogen dose (50N, 100N, 150N and 200N, N=kg available N ha^{-1}) for compost, anaerobic and pellet of sewage sludge, per year with crop extraction. 86/278/EEC: Soil threshold for concentration of Zn in soil (kg ha^{-1}) in Directive 86/278/EEC (EU, 1986). EUDWD: Soil threshold for concentration of Zn in soil (kg ha^{-1}) in EUDWD (EC, 2000). S1: Soil threshold for concentration of Zn in soil (kg ha^{-1}) in Final Report in option Moderate changes (S1) (EC, 2008). S2: Soil threshold for concentration of Zn in soil (kg ha^{-1}) in Final Report in option More significant changes (S2) (EC, 2008).



Capítulo 3. Circular economy: using lime stabilised bio-waste based fertilisers to improve soil fertility in acidic grasslands

Abstract

The Circular Economy Package adopted by EU proposed new rules on the bio-waste based fertilisers that reducing waste, energy consumption and environmental damage. Therefore, it is important to characterize the effect of these fertilisers on soil fertility and pasture/crops growth. The aim of this study was to evaluate the effect of different doses of bio-waste based fertilisers with amendment value stabilized and sanitized with lime on the chemical soil properties, pasture production and its botanical composition compared with control conventional treatments (no fertilisation and mineral fertilisation) combined with lime and without lime in pastures established in Galician acid soils. The results of this experiment show that the high doses of the bio-waste based fertilisers improved the soil fertility and therefore increased the pasture production and modified its botanical composition which was more sensible to the variation of soil fertility than the production. Therefore, the botanical composition could be considered as a good bio-indicator of changes in soil fertility when the bio-waste based fertilisers are used in the agriculture. Moreover, the soil and pasture results associated to the organic amendments were similar to the results obtained when the conventional practices of the area were carried out such as the mineral fertilization. Therefore, the partial or total substitution of mineral fertilizers by bio-waste based fertilisers could be a viable alternative to reduce cost of production in the farms and the environment impact from waste and chemical fertilisers.

1. Introduction

The valorisation of organic waste as amendments for agricultural use is an effective way to recycle organic matter and nutrients contained therein (Steven, 2011; Scotti et al., 2015; Katheem et al. 2016). In 2015, the European Commission (EC) adopted an ambitious Circular Economy Package, which includes revised legislative proposals on waste to stimulate Europe's transition towards a circular economy which will boost global competitiveness, foster sustainable economic growth and generate new jobs (EC, 2015).

One of the main EC proposals to use waste as fertilisers is to establish common rules on fertilisers as diverging rules and standards hamper the manufacturing of organic and waste based fertilisers from inputs such as food waste or manure. In 2016, the EC revised the European Union (EU) regulation on fertilisers to develop a EU-wide market for bio-nutrients while ensuring safety and quality of the EU fertilisers (EC, 2016). Waste should be stabilised and sanitized to avoid undesired effects (EC, 2001; Arthurson, 2008). These processes change the waste characteristics and its agricultural value (Czechowski and Marcinkowski, 2006). Alkaline stabilization with lime is an advanced treatment (EC, 2001) for organic waste that is regarded as an attractive alternative to aerobic or anaerobic digestion (Czechowski and Marcinkowski, 2006), although nowadays its use is not extended in the EU (Werle and Wilk 2010). In the EU, the waste generation, excluding major mineral waste (2004–2012), was 4.3% from agriculture, forestry and fishing and 19.6% from waste water (Eurostat yearbook, 2016).

In Galicia (NW Spain), food industry as well as primary (agriculture, forestry and fishing) and water/waste activities represent over 11% of the gross domestic product (GDP). The primary sector represents over 18% of total industries in Galicia in 2014. Arables, grasslands and forests represent 12.9%, 15.1% and 60.9% of total areas in Galicia, respectively (IGE, 2016). In this region, 2.2 millions of tonnes of Non-Harzous Waste (NHW) were generated from industry in 2014 from which 82% was recycled and re-used (Xunta de Galicia, 2016) in recovery operations such as the established in the

Annex II of the Spanish and EU regulations dealing with waste (EC, 2008; BOE, 2011). Around 25.8% of total NHW have the potential to be used as an organic resource for the development of fertilisers or substrates, distributed in sewage sludge (8.5%), bio-waste (8.7%), wood ash (7.8%) and manure (0.8%).

On the other hand, in Galicia, the soil acidity is a natural phenomenon derived from its humid climate, the prevalence of subtractive systems, frequent fires and, often, acidic parent material (Álvarez et al., 2009) which implies a low soil fertility for arable crops with an excess of aluminium in the soil (Mombiela and Mateo 1984). Therefore, it is advisable to perform management activities such as liming and fertilisation to increase the soil fertility and to neutralise acidity which can limit pasture production (Rigueiro-Rodríguez et al., 2008). One option adopted in many countries around the world is the use of organic waste as fertiliser (EPA, 1994) because of its low cost compared with mineral fertiliser (EFMA, 2009) and because it recycles nutrients with increasing low market availability (Sigua et al., 2005; Mosquera-Losada et al., 2010). Moreover, the use of lime to stabilise the bio-waste based fertilisers improves the amendment value of the fertilisers which is very appropriate for the Galician acid soils.

The aim of this study was to evaluate the effect of different doses of bio-waste based fertilisers with amendment value stabilized and sanitized with lime on the chemical soil properties, pasture production and its botanical composition compared with control conventional treatments (no fertilisation and mineral fertilisation) combined with lime and without lime in pastures established in Galician acid soils.

2. Materials and methods

2.1. Characteristics of the study site

The experiment was conducted in Curtis (Lugo, Galicia, north-western Spain, European Atlantic Biogeographic Region) at an altitude of 450 m a.s.l. The mean monthly temperature and precipitation for the period 2011–2014 as well as for the last historic period of 25 years (1985-2010) are shown in Figure 1.

The data shows that 2011 was the driest year, with annual precipitation levels (966.4 mm) lower than the annual mean for the study area (1069 mm). In 2012, 2013 and 2014, the total annual rainfall (1179.9 mm, 1744.1 mm and 1583.6 mm, respectively) was higher than the mean precipitation over the 25 years due to the especially high precipitation levels in the last months of 2012 and at the beginning and at the end of 2013 and 2014. However, in these years, the drought periods were also registered during July and August which could have been unfavourable for pasture production. The annual mean temperatures in 2011 (11.0 ºC), 2012 (10.2 ºC), 2013 (10.0 ºC) and 2014 (10.5 ºC) were lower than that annual mean for the 25-year period (12 ºC).

At the beginning of the experiment the soil texture was sandy loam (68.3% sand, 22.8% silt and 5.8% clay). The water soil pH (4.86) and the effective cation exchange capacity (ECEC) (8.3 cmol(+) kg-1) were low and the saturation percentage of aluminium was high (76,02%). Moreover, all total heavy metal concentrations in the soil (Table 1) were below the maximum threshold for using sewage sludge fertiliser as specified by the European Union Directive 86/278/EEC (EU, 1986) and Spanish legislation under R.D.1310/1990 (BOE, 1990).

2.2. Experimental design

An experiment with a randomised block design was performed in 48 experimental plots (twelve treatments × four replicates) of 8 m^2 (2 m x 4 m). Each plot was sown in the spring of 2011 with a mixture of 25 kg ha^{-1} of *Lolium perenne* var. Brigantia and 5 kg ha^{-1} of *Trifolium repens* cv. Huia after shrub clearing and ploughing. The established treatments were four doses (T1:1 Mg ha^{-1}, T2.5: 2.5 Mg ha^{-1}, T5: 5 Mg ha^{-1} and T10:10 Mg ha^{-1}) per type of bio-waste based fertiliser (HP: bio-waste based fertiliser with high pH (5:6:17:28, N:P_2O_5:K_2O:Ca) and LP bio-waste based fertiliser with low pH (11:12:7:6, N:P_2O_5:K_2O:Ca)) combined with mineral fertilisation (MIN: 500 kg ha^{-1} 8:24:16 (N:P_2O_5:K_2O)) before sowing. These doses of HP and LP were selected to replicate the same effects of the doses of fertilisation and liming which are usually applied in the area. Moreover, four control treatments were also established: no fertilisation (NF) and mineral fertilisation (MIN: 500 kg ha^{-1} 8:24:16 (N:P_2O_5:K_2O)) combined with (+lime: 2.5 Mg $CaCO_3$ ha^{-1}) or without liming (-lime) applied before sowing; which represents conventional agricultural practices carried out in the area.

2.3. Bio-waste based fertilisers

The bio-waste based fertilisers used in this experiment have been stabilized with lime (advanced treatment (EC 2001)) or wood ash, using two criteria in order to evaluate the effect on nutrient release and therefore their fertiliser value in acid soils. The stabilisation with lime increased the amendment value of the bio-waste based fertilisers of this study. One of the bio-waste based fertiliser (LP) was composed by urban sewage sludge (65%), chicken manure (25%) and wood ash (10%). The other bio-waste based fertiliser (HP) was composed by urban sewage sludge (35%), animal by-product (20%), quicklime (18%), wood kindling (17%) and wood ash (10%). The first bio-waste based fertiliser (LP) had a lower pH in water (6.97) than the second one (HP) (13.19) due to the different proportion of the used waste. The materials presented in the composition of these bio-waste based fertilisers are included into the NHW generated in Galicia with high potential to be used in the manufacture of fertilisers (amendments) (BOE, 2013) or substrates (BOE, 2010). The composition of both bio-waste based fertilisers applied in this study is summarised in Table 2. The concentration of main nutrients, N and P were higher in LP than in HP due to the dilution effect by the higher addition of lime in the HP bio-waste fertiliser. However, the concentrations of Ca, Mg, Na and K were higher in HP than in LP probably because usually these cations are part of lime in large proportions. The total heavy metals concentrations in both bio-waste based fertilisers fulfil the limits established in the Spanish regulation for fertilisers (BOE, 2013).

2.4. Field samplings and laboratory analyses

A composite soil sample per plot was randomly taken at a depth of 25 cm in March 2012, February 2013 and February 2014 as described in R.D. 1310/1990 (BOE, 1990). In the laboratory, the soil samples were air-dried, passed through a 2 mm sieve and ground with an agate mortar. The soil pH was determined in water (1:2.5) (Faithfull, 2002), and an extraction with 0.6 N $BaCl_2$ was used to determine the concentrations of Al and the exchangeable cations (K, Ca, Mg and Na) in the exchange complex (Mosquera and Mombiela, 1986). The exchangeable K, Ca, Mg and Na concentrations were measured with a VARIAN 220FS spectrophotometer using the atomic emissions for K and Na and the absorptions for Ca and Mg (VARIAN, 1989). The Al concentration was analysed after valoration with 0.01 N NaOH using phenolphthalein (1%) in an alcohol-based solution as an indicator (Mosquera and Mombiela, 1986). The effective cation exchange capacity (ECEC) was determined by taking the sum of K + Ca + Mg + Na + Al and the saturation percentage of Al, K, Ca, Mg and Na using the quotients Al/ECEC, K/ECEC, Ca/ECEC Mg/ECEC and Na/ECEC, respectively (Mosquera and Mombiela, 1986).

In order to determine the pasture production in each plot, the pasture was harvested using a hand harvester in June, August and December 2012 and in July and December 2013, as is conventional for the area, when the pasture height reached about 20 cm.

Fresh pasture was weighed in situ and a representative subsample was taken to the laboratory. Once in the laboratory, two subsamples (95-105 g each) were dried (72 hours at 60ºC) and weighed to estimate dry matter production. Moreover, one of the two subsamples was separated by hand according to the different species and senescent material to determine its composition on a dry weight basis and use to estimate different biodiversity variables (alpha-biodiversity, and abundance diagrams). The cumulative production of the pasture was calculated by summing the consecutive harvests.

Total number of species (i.e. annual alpha-biodiversity) and annual abundance diagrams (Magurran, 1988) were analysed. In the annual abundance diagrams the percentage of senescent material was not taken into account.

2.5. Statistical analysis

The soil variables were analysed with repeated analysis of variance (ANOVA) measures (PROC GLM procedure), and Mauchly's criterion was used to test for sphericity. If the sphericity assumption was met, univariate approach output was used, otherwise multivaritae output (Wilks' Lambda test) was taken into account. The model used was $Y_{ij} = \mu + A_i + T_j + TA_{ji} + \varepsilon_{ij}$, where Y_{ij} is the dependent variable, μ is the variable mean, A_i is the year i, T_j indicates treatment j, TA_{ji} is the treatment–year interaction and ε_{ij} is the error.

The data obtained from the soil in each year and pasture (production and species richness) were also analysed by ANOVA (PROC GLM procedure) using the model $Y_{ik} = \mu + T_i + B_k + \varepsilon_{ik}$, where Y_{ik} is the dependent variables, μ is the variable mean, T_i indicates treatment i, B_k in the block k and ε_{ik} is the error.

The LSD test was used for subsequent pairwise comparisons ($p < 0.05$; a = 0.05) if the ANOVA was significant. The statistical soft- ware package SAS (2001) was used for all analyses.

3. Results

3.1. Soil

3.1.1. Water soil pH

In this study, the soil pH was higher in 2013 compared with 2012 and 2014 ($p<0.01$) (Table 3). In 2012 and 2014, water soil pH (Figure 2) was significantly higher in the control treatments combined with lime (+lime) ($p<0.05$) than without lime (-lime). High dose of HP (T10) generally increased more the water soil pH than the other doses of HP and the doses of LP. Moreover, the soil pH was significantly higher when the high doses (T5 and T10) of HP were applied compared with the control treatments (NF and MIN) without lime (-lime) ($p<0.001$).

3.1.2. ECEC and saturation percentages of Al, K, Ca, Mg and Na in the soil exchange complex

Table 3 shows that the soil ECEC and the saturation percentages of K and Na increased in 2013 compared with 2012 and 2014 ($p<0.001$). However, the saturation percentage of Ca was lower in 2013 than in the other years of the study. The saturation percentage of Mg decreased in 2014 compared with 2012 and 2013 and the saturation percentage of Al increased over time.

In 2012, it was observed a positive effect of liming (+lime) on ECEC in the NF treatment ($p<0.001$) (Figure 3). Moreover, in general, the high dose of HP (T10) increased more the soil ECEC than the other doses of HP and the doses of LP in 2012 and 2013. In these years, the ECEC was also significantly higher when the high dose (T10) of HP was applied compared with the control treatments (NF and MIN) without lime (-lime).

The saturation percentages of the different cations (Al, Mg, Ca, Na and K) in the soil ECEC can be seen in Figure 4 ($p<0.001$). In the control treatments (NF and MIN), the saturation percentage of Ca was positively affected by liming (+lime) during the whole experiment, being the effect of liming the opposite when the saturation percentage of Al was evaluated. Moreover, it was observed a negative effect of liming (+lime) on the saturation percentage of K in the NF treatment in 2012 and in the MIN treatment in 2013 and 2014, on the saturation percentage of Mg in the MIN treatment in 2012 and 2013 and on the saturation percentage of Na in the NF and MIN treatments in 2012 and 2013. However, in 2014, in the NF treatment, the saturation percentage of Mg increased when lime was applied into the soil (+lime).

During the three years of the study, the highest dose (T10) of HP generally increased more the saturation percentage of Ca than the other doses of HP and LP and therefore this treatment (T10 dose of HP) reduced the saturation percentage of Al ($p<0.001$). The saturation percentage of Mg was also higher when the T10 dose of HP was applied compared with LP in 2014. However, the saturation percentages of K and Na decreased more with the T10 dose of HP than with LP in 2012 and 2013.

Finally, the high doses (T5 and T10) of HP increased more the saturation percentage of Ca than the control treatments (NF and MIN) without liming (-lime), being the opposite result when the saturation percentage of Al was evaluated ($p<0.001$). Moreover, it was also observed a negative effect of the highest dose (T10) of HP on the saturation percentage of K and Na in 2012 and 2013 and in general on the saturation percentage of Mg compared with the control treatments (NF and MIN) without lime (-lime) during all years of the study.

3.2. Pasture

3.2.1. Pasture production

Pasture production was significantly higher in 2012 than in 2013 (2012: 5.29^a and 2013: 3.26^b; expressed as Mg ha^{-1}; different superscript letters indicate significant differences between years) ($p<0.001$).

Figure 5 shows that pasture production was significantly reduced in the NF treatment with or without lime in 2012 ($p<0.001$) and 2013 ($p<0.001$) compared with the mineral treatment (MIN). In the mineral treatment (MIN), the addition of lime (+lime) decreased pasture production in 2013 compared with the treatment without lime (-lime). In general, in 2012, the production of pasture was significantly higher in the highest dose (T10) of LP compared with the rest of doses of LP and HP. Finally, in 2012 and 2013, the bio-waste based fertilisers (LP and HP) increased the pasture production compared with no fertilisation treatments (NF).

3.2.2. Species richness

From the list of species (Table 4), it can be seen that 41 species were found during the two years of study, belonging to 14 different families, of which 15 represented the family of the Poaceae, six of the Fabaceae, six of the Asteraceae, three of the Ericaceae, two of the Rosaceae, leaving just one representative for each of the following families: Boraginaceae, Bryophyta s.s., Campanulaceae, Caryophyllaceae, Dennstaedtiaceae, Polygonaceae, Rubiaceae, Scrophulariaceae and Violaceae. Out of the 41 species, 7 were woody and 33 herbaceous species. The total number of species identified was 36 and 40, for the years 2012 and 2013, respectively. Of the total, 35 species were present in the plots throughout the whole period evaluated (84% of the total identified). Out these 35 species, 7 were woody and perennials, 18 were herbaceous and perennials and 12 were herbaceous and annuals with the exception of *Digitalis purpurea* L. that is a biannual species. Out the 42 species found in this experiment, one appeared at the beginning of the experiment but disappear over time (*Vicia* sp.), and another 5 appeared later (*Agrostis curtisii* Kerguelen, *Venula sulcata* (Gay ex Boiss) Dumort, *Calluna vulgaris* (L.) Hull, *Moss* and *Poa annua* L.), but were then present until the end.

On the other hand, the mean number of species per treatment and year was significantly lower in 2013 than in 2012 (2012: 12a and 2013: 9b; different superscript letters indicate significant differences between years) (p <0.001). In both years, the MIN treatment combined with lime (+lime) generally reduced de number of species compared with the MIN treatment without lime (-lime) and with the NF treatment with lime (+lime) and without lime (-lime) (Figure 6) (p <0.001). Moreover, in 2012, the number of species decreased more with the high doses (T10) of LP and HP than with the low doses (T1 and T2.5) of both bio-waste based fertilisers. The negative effect of the high doses (T10) of LP on the number species it was also observed in 2013 compared with the low doses (T1) of LP and HP. Moreover, in both years of the experiment, the number of species was lower when the high dose (T10) of HP was applied to the soil compared with the NF treatment.

Finally, the abundance diagrams (Figures 7 and 8) show that, in general, throughout the study, *Agrostis capillaris* L. was the dominant species in all treatments (almost 60% in the control treatments) with the exception in 2012 of those treatments with the highest dose (T10) of bio-waste based fertilisers (LP and HP) in which *Agrostis capillaris* L. shared dominance with the legume *Trifolium repens* L.

4. Discussion

This study was carried out in an acid soil of Galicia (NW Spain). In this region, the soil acidity is a natural phenomenon derived largely from its humid climate, with higher rainfall than evapotranspiration during most of the year (Álvarez et al., 2009). In this experiment, it was observed an increase of soil pH, ECEC and saturation percentages of

K and Na at the beginning of 2013 compared with 2012, being this increase very slight in the case of the soil pH. This result could be due to the application of the bio-waste based fertilisers to the soil when the experiment was established (2011). Authors as Diacono et al. (2010) have shown in their studies that the regular addition of bio-waste based fertilisers to the soil increases the soil physical fertility, mainly due to the improvement of the soil aggregates stability. The application of bio-waste based fertilisers to the soil can also enhance the availability of K, the extractable P, the organic carbon and the soil organic N content without raising the nitrate leaching to groundwater (Diacono et al., 2010). Moreover, the improvement of these soil variables at the beginning of 2013 compared with 2012 could be also explained by an increase in the mineralisation rate of soil organic matter due to (i) the application of mineral fertiliser in all plots and (ii) the increase of soil moisture derived from the higher levels of rainfall in 2012 (1179.9 mm) than in 2011 (966.4 mm). Soil moisture is one of the major factors controlling mineralisation and this ecological factor is key in the regulation of the soil nutrient cycling (EPA, 1994). Therefore, the increase in the mineralisation rate might have favoured the release of nutrients into the soil, which improved the ECEC. Similar results were previously described by Ferreiro-Domínguez et al. (2014) in a silvopastoral system established in the same region and fertilised with different doses of sewage sludge (50 and 100 kg total N ha^{-1}) combined with liming. Moreover, the higher saturation percentages of K and Na at the beginning of 2013 than in 2012 are explained by the addition of K inorganic from mineral fertiliser and the easiness of Na release from the bio-waste based fertiliser. The increase of K and Na probably explains that the saturation percentage of Ca decreased at the beginning of 2013 compared with 2012 because this cation is characterized by its strong antagonism with the K (Barber, 1995, Mosquera-Losada et al., 2012) and the Na (Kopittke, 2012).

However, at the beginning of 2014, soil pH, ECEC and saturation percentages of K, Mg and Na decreased compared with February 2013 which could be explained by several factors. Firstly, the high precipitation recorded during 2013 (1744.1 mm), mainly in the last months of the year, could have favoured the leaching of the cations through the soil profile, being the solubility order of ECEC cations $Na^+ > K^+ > Mg^+ > Ca^{2+} > Al^{3+}$ (Calvo et al., 1987; Álvarez et al., 1992). Secondly, because the bio-waste based fertilisers were only applied at the beginning of the study, suggesting that cations could have been extracted by the pasture (Adams et al., 2001). The soil cations extraction by pasture could also explain the low saturation percentage of Ca at the end of the experiment as pasture has higher levels of Ca than K and Na and the subsequent increase of the saturation percentage of Al over time, mainly observed in the treatments without liming and bio-waste based fertilisers. These results were previously described by other authors as Ferreiro-Domínguez et al. (2011) and Mosquera-Losada et al (2012) in silvopastoral systems established in acidic soils of Galicia under *Pinus radiata* D. Don.

On the other hand, the improvement of soil fertility caused by the bio-waste based fertilisers inputs to the soil probably increased the pasture production in 2012. However, the residual effect of the improvement of the soil fertility found in February 2013 did not affect the pasture production of 2013. The changes in the soil fertility probably also explain that the number of species was higher in 2013 (40 species) than in 2012 (36 species). The less restrictive edaphic conditions could have led to the establishment of gramineous species such as *Agrostis curtisii* Kerguelen, *Avenula sulcata* (Gay ex Boiss) Dumort and *Poa annua* L. in 2013. Moreover, the higher rainfall in 2013 than in the other years of the experiment probably favoured the establishment of Moss in that year. These results demonstrate that the botanical composition of the pasture was more sensible to the variation of soil fertility caused by the application of bio-waste based fertilisers to the soil than the pasture production. Therefore, the botanical composition could be considered as a good bio-indicator of changes in soil fertility when the bio-waste based fertilisers are used in the agriculture. The positive effect of the organic fertiliser on pasture and composition of the pasture was previously observed by several authors in the same area (Mosquera-Losada et al., 2009; Ferreiro-Domínguez et al., 2011). However, the mean number of species per treatment and year decreased over time which could be explained by a reordering of the species due to the change of dominance.

The comparison of the effect of the application of lime and the different doses of bio-waste based fertilisers on the soil variables showed a positive effect of liming and the high doses of HP (T10) on soil pH, ECEC and saturation percentage of Ca and also on the saturation percentage of Mg. The positive effect of lime on these soil variables could be due to the increase of the exchangeable Ca (Slattery et al., 1995), which tends to improve the physical and chemical properties of the soil and microbial activity (Baley, 1995; Álvarez et al., 1992), causing organic mineralisation and therefore, nutrient release (Wheeler, 1998). Similar results were also found in experiments carried out in the same area in which it was evaluated the effect of lime on pasture production and *Pinus radiata* D. Don growth (Ferreiro-Domínguez et al., 2014). In the case of the bio-waste based fertilisers, the improvement of the soil pH, the ECEC and the saturation percentages of Ca and Mg when the high doses of HP (T10) were applied to the soil compared with the other doses of HP and LP could be explained by the higher pH of HP (13.19) than LP (6.96). Moreover, the high dose of HP (T10) implied higher inputs of Ca and Mg in the soil (2774 kg Ca ha^{-1} and 200 kg Mg ha^{-1}) than the other doses of HP and the other doses of LP (T10: 549 kg Ca ha^{-1} and 41 kg Mg ha^{-1}). On the other hand, the increase in the saturation percentage of Ca after the application of lime and high doses of HP decreased the saturation percentages of Al, K, Mg and Na due to the antagonism between these cations and Ca captured by the ECEC positions (Keltjens and Tan, 1993; Smith, 1996; Prasad and Power, 1997; Fageria, 2001) and the stronger energy of adsorption of Ca than Mg, K or Na (Foth, 1990). Finally, the soil fertility was generally similar in the plots which received high doses of HP compared with the plots in which it was carried out conventional management practices of the area such as the liming or the mineral fertilisation combined with lime. It is important to be aware that HP, as in case of lime-stabilized biosolids, has a liming equivalency and effectiveness lower than lime ($CaCO_3$) (Sloan and Basta, 1995; Guo et al., 2012) and for this reason it is necessary to apply higher doses of HP than lime to increase soil pH, ECEC and saturation percentage of Ca.

In this experiment, it was observed a clear effect of the established treatments on the production and the botanical composition of the pasture. In general, the mineral fertilisation increased pasture production compared with the NF treatment in which the soil chemical properties remaining to be very poor (low ECEC and high saturation percentage of Al). The positive effect of the mineral fertilisation on pasture production was previously described by numerous authors (López-Díaz et al., 2007; Courtney and Mullen, 2008; Mosquera-Losada, et al. 2012; 2016) and could be explained by the inputs of inorganic N to the soil which reduces the C/N ratio that increases the mineralisation of soil organic matter and therefore the availability of cations for the pasture (Whitehead, 1995). On the other hand, in the first year of the study, the pasture production was higher when the high dose (T10) of LP was applied to the soil compared with the lower doses of LP and HP probably because the high dose of LP implied a higher inputs of N in to the soil (101 kg N ha^{-1}) that the other doses of LP and HP (T5:

24 kg N ha^{-1}), being the N the main limiting nutrient for plants growth (Burgos et al., 2016; Obriot et al., 2016) but also because the higher dose of HP caused a higher release of nutrients from soil as the mineralization was activated. Moreover, in general, in both years of the experiment, the pasture production associated to the low and intermediate doses (T1, T2.5 and T5) of the bio-waste based fertilisers was similar to the pasture production obtained in the mineral treatment which is indicative of the nutrient release from the soil organic matter of the area of this experiment (López-Díaz et al., 2007; Mosquera-Losada et al., 2012). Other authors as Herencia et al (2007) also found in their studies that the use of organic fertilisers improves soil fertility and produces similar yields and nutrient composition in the crops compared with the mineral fertilisation. In any case, it is important to be aware that when the high doses of the HP and LP bio-waste based fertilisers were applied to the soil, pasture production obtained in this study in 2012 was within the range of pasture production found in more than 80% of the Galicia plots (6.25-18,75Mg ha-1) MAPAMA (2013). However, pasture production of this experiment was lower that the range of pasture production established by MAPAMA (2013) when the other doses of the bio-waste based fertilisers and the NF and MIN treatments were applied in 2012 and in all treatments in 2013. These results indicate, firstly, the positive effect of the application of high doses of these bio-waste based fertilisers on pasture production compared with the conventional management practices carried out in the area (mineral fertilisation) and, secondly, that the positive effect of the bio-waste based fertilisers on pasture production decrease over time and therefore it is advisable its application periodically. On the other hand, the higher inputs of N to the soil associated to the mineral treatment and the high doses of HP and LP probably decreased the number of species in these treatments compared with the low doses of the bio-waste based fertilisers and the NF treatment. Other authors as

Willems et al. (1993), Hyvonen and Salonen (2002) or Dise and Stevens (2005) also found in their studies that the inputs of nutrients to the soil disfavour biodiversity although this effect depends on several factors such as the initial soil conditions or the capacity of adaptation of the species to perturbations in a determined edaphoclimatic context. In addition to the effect of the bio-waste based fertilisers doses on the total number of species, this treatment also altered the species distribution with regard to their relative proportion. In general, *Agrostis capillaris* L. was the most abundant species in all treatment of the study in 2012 and 2013 with the exception in 2012 of those treatments with the highest dose (T10) of bio-waste based fertiliser (LP and HP) in which *Agrostis capillaris* L. shared dominance with *Trifolium repens* L. The change in the proportion of *Trifolium repens* L. due to the application of high doses of HP and LP could explained by the improvement of soil fertility associated to these treatments because this species requires more fertile soils than other spontaneous species as *Agrostis capillaris* L. (Grime et al., 2007), and is very sensible to grow up in soils with a high level of saturation Al. Other authors as Mosquera-Losada et al. (2009) and Ferreiro-Domínguez et al. (2011) in studies carried out in the same area also found that the number of species and the proportion of the species in the pasture were modified by the management practices such as organic fertilisation and the liming. However, in any case, it is important that these management practices are compatible with the conservation of the species that are characteristic of the acid soils of the Atlantic zone to maintain the vegetal biodiversity.

5. Conclusion

The use of bio-waste based fertilisers in the agriculture is a good option compared with the conventional management practices in the area such as the liming and the mineral fertilisation due to the bio-waste based fertilisers imply a nutrient recycling at the same time that the soil fertilisation is improved. In this study, the improvement of the soil fertility associated to the application of bio-waste based fertilisers increased the pasture production and modified its botanical composition, mainly when high doses of the bio-waste based fertilisers were applied. The botanical composition of pasture was more sensible to the variation of soil fertility caused by the bio-waste based fertilisers than the pasture production and therefore the botanical composition could be considered as a good bio-indicator of changes in soil fertility when the bio-waste based fertilisers are applied to the soil. Moreover, the soil and pasture results associated to the bio-waste organic fertilisers were similar to the results obtained when the conventional practices of the area were carried out. Therefore, the partial or total substitution of mineral fertilisers by bio-waste based fertilisers could be a viable alternative to reduce cost of production in the farms and the environment impact from waste and chemical fertilisers.

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Table captions

Table 1. Heavy metal concentrations in the soil at the beginning of the experiment and the legal limits established by European Directive 86/278 (EU, 1986) and Spain R.D. 1310/1990 (BOE, 1990). Limits depend on soil pH (minimum: soil pH< 7; maximum: soil pH> 7). A dash (–) signifies an element concentration below the detection limit of the technique used for its determination.

	Heavy metal concentrations (mg kg^{-1})					
	Zn	Cu	Cr	Cd	Ni	Pb
Initial soil	27.5	17.7	8.9	-	11.6	14.7
Spanish legal limits	150-450	50-210	100-150	1-3	30-112	50-300

Table 2. Chemical properties of the organic amendment with Low pH (LP) and High pH (HP), the limits (F) established in the Spanish regulation for fertilisers (BOE, 2013) and inputs for high dose (T10) of both amendments (LP, HP).

Parameter	LP	HP	F	LP T10 (kg ha^{-1})	HP T10 (kg ha^{-1})
pH (water)	6.96	13.19	-	-	-
DM (% dry matter)	35,8	73,1	-	-	-
N (%)	2.82	0.66	-	101	48
P (%)	1.41	0.37	-	51	27
K (%)	1.61	1.91	-	58	140
Na (%)	0.41	1.06	-	15	78
Ca (%)	15.33	37.94	-	549	2774
Mg (%)	1.14	2.73	-	41	200
Cd (mg kg^{-1})	1.5	<0.1	3	0.005	<0.001
Ni (mg kg^{-1})	32.7	62.1	100	0.117	0.454
Pb(mg kg^{-1})	34.6	16.2	200	0.124	0.118
Zn (mg kg^{-1})	310.6	148.0	1000	1.112	1.082
Hg (mg kg^{-1})	0.120	0.112	2.5	<0.001	<0.001
Cr (mg kg^{-1})	8.7	13.1	300	0.031	0.096
Cu (mg kg^{-1})	123.8	41.0	400	0.443	0.300

Table 3. Water soil pH, effective exchange capacity (EEC) (cmol (+) kg^{-1}) and saturation percentage of Al, K, Ca, Mg and Na in the soil exchange complex (%) in 2012, 2013, and 2014. Different letters indicate significant differences between years ($p<0.001$). SEM = standard error of the mean.

Parameter	Year			SEM
	2012	2013	2014	
pH (H_2O)	4.95 b	5.07 a	4.95 b	0.02
ECEC (cmol(+) kg^{-1})	9.16 b	10.15 a	6.02 c	0.23
Al (%)	37.68 c	43.27 b	49.67 a	1.16
Ca (%)	37.50 a	25.42 c	31.43 b	1.24
Mg (%)	10.09 a	10.00 a	6.61 b	0.20
Na (%)	13.79 b	16.54 a	9.63 c	0.36
K (%)	0.93 c	4.77 a	2.67 b	0.14

Table 4. Description of the species found in the botanical analysis. A: annual; P: perennial; SP: species type; H: herbaceous; L: woody. X: indicates that the species appeared in the year above the column. O: indicates that the species did not appear in the year above the column**.**

Code	Family	Species	2012	2013	A/P	SP
Acm	Asteraceae	*Achillea millefolium* L.	X	X	P	H
Card	Asteraceae	*Carduus* sp.	X	X	A	H
Mtr	Asteraceae	*Matricaria chamomilla* L.	X	X	A	H
Sen	Asteraceae	*Senecio vulgaris* L.	X	X	A	H
Snc	Asteraceae	*Sonchus asper* L (Hill)	X	X	A	H
Tar	Asteraceae	*Taraxacum officinale* Weber	X	X	P	H
Lit	Boraginaceae	*Glandora prostrata* (Loisel.) DC	X	X	P	L
Mus	Bryophyta s.s.	*Moss*	O	X	-	-
Jas	Campanulaceae	*Jasione montana* L.	X	X	A	H
Ste	Caryophyllaceae	*Stellaria media* L. (Vill)	X	X	A	H
Pt	Dennstaedtiaceae	*Pteridium aquilinum* (L.) Kuhn:	X	X	P	H
Call	Ericaceae	*Calluna vulgaris* (L.) Hull	O	X	P	L
Dab	Ericaceae	*Daboecia cantabrica* (Hudson) C. Koch	X	X	P	L
Erc	Ericaceae	*Erica cinerea* L.	X	X	P	L
Cyt	Fabaceae	*Cytisus striatus* (Hill) Rothm.	X	X	P	L
Lc	Fabaceae	*Lotus corniculatus*	X	X	P	H
Orn	Fabaceae	*Ornithopus compressus* L.	X	X	A	H
Tri	Fabaceae	*Trifolium repens* L.	X	X	P	H
Ulx	Fabaceae	*Ulex europaeus* L.	X	X	P	L
Vci	Fabaceae	*Vicia* sp.	X	O	A	H
Agca	Poaceae	*Agrostis capillaris* L. = Agrostis tenuis	X	X	P	H
Agu	Poaceae	*Agrostis curtisii* Kerguelen	O	X	P	H
Agdu	Poaceae	*Agrostis durieui* Boiss. & Reut. ex Willk.	X	X	P	H
Atx	Poaceae	*Anthoxantum odoratum* L.	X	X	P	H
Avn	Poaceae	*Avenula sulcata* (Gay ex Boiss) Dumort	O	X	P	H
Brd	Poaceae	*Bromus diandrus* Roth.	X	X	A	H
Brm	Poaceae	*Bromus hordeaceus* L.	X	X	A	H
Dg	Poaceae	*Dactylis glomerata* L.	X	X	P	H
Dat	Poaceae	*Danthonia decumbens* (L.) DC	X	X	P	H
Hl	Poaceae	*Holcus lanatus* L.	X	X	P	H
Hm	Poaceae	*Holcus mollis* L.	X	X	P	H
Lp	Poaceae	*Lolium perenne* L.	X	X	P	H
Mol	Poaceae	*Molinia caerulea* (L.) Moench	X	X	P	H
Poa	Poaceae	*Poa annua* L.	O	X	A	H
Psu	Poaceae	*Pseudarrhenatherum longuifolium* (Thore) Rouy	X	X	P	H
Rma	Polygonaceae	*Rumex acetosella* L.	X	X	P	H
Pot	Rosaceae	*Potentilla erecta* (L.) Raeusch	X	X	P	H
Rub	Rosaceae	*Rubus* spp.	X	X	P	L
Gal	Rubiaceae	*Galium aparine* L.	X	X	A	H
Dp	Scrophulariaceae	*Digitalis purpurea* L.	X	X	B	H
Vol	Violaceae	*Viola* sp.	X	X	P	H

Figure captions

Figure 1. Monthly precipitation and mean temperatures for the study area for the years 2011–2014 and mean data for the last 25 years (1985-2010). T: mean monthly temperature (ºC); T25: mean temperature over the last historic period of 25 years (ºC); P: monthly precipitation (mm) and P25: mean precipitation over the last historic period of 25 years (mm).

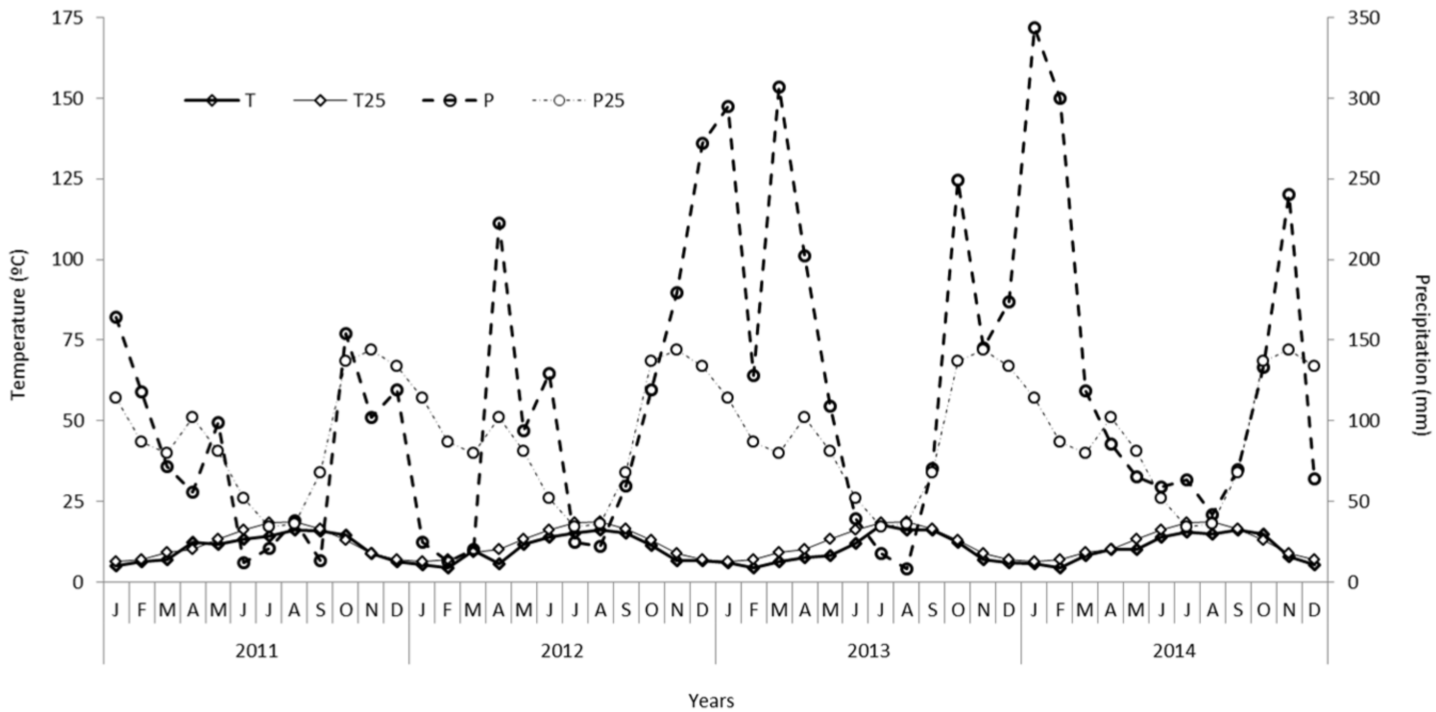


Figure 2. Water soil pH under each treatment in 2012, 2013 and 2014. HP: bio-waste based fertiliser with high pH, LP: bio-waste based fertiliser with low pH, T1, T2.5, T5, T10: bio-waste based fertilisers doses (1, 2.5, 5 and 10 Mg ha^{-1}) combined with mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O)), NF: no fertiliser, MIN: mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O) with lime (2.5 Mg CaCO3 $ha^{-1)}$ (+lime) and without lime (-lime). Different letters indicate significant differences between treatments within the same year. Bars indicate the standard error of the mean.

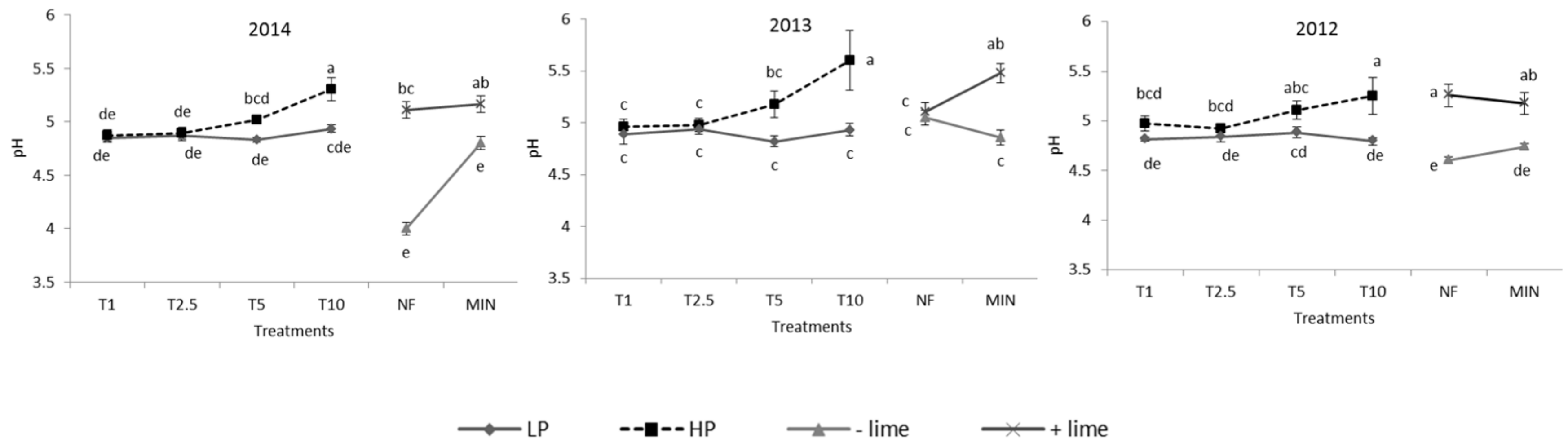


Figure 3. Soil effective exchange capacity (EEC) (cmol (+) kg^{-1}) under each treatment in 2012, 2013 and 2014. HP: bio-waste based fertiliser with high pH, LP: bio-waste based fertiliser with low pH, T1, T2.5, T5, T10: bio-waste based fertilisers doses (1, 2.5, 5 and 10 Mg ha^{-1}) combined with mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O)), NF: no fertiliser, MIN: mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O) with lime (2.5 Mg CaCO3 ha^{-1}) (+lime) and without lime (-lime). Different letters indicate significant differences between treatments within the same year and treatments are no significantly different if no letters are shown. Bars indicate the standard error of the mean.

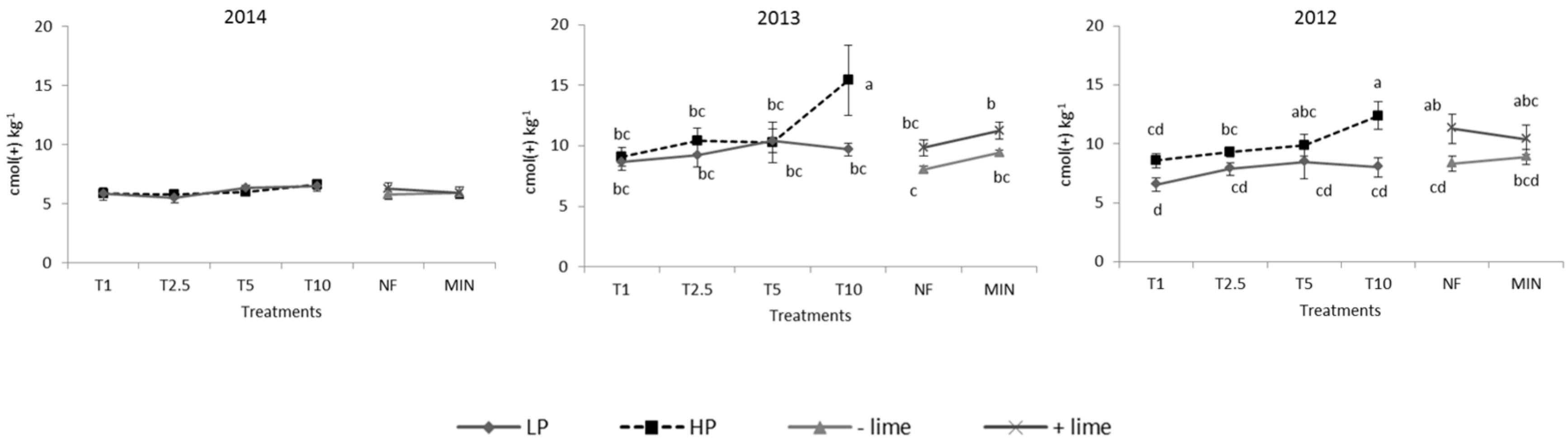


Figure 4. Saturation percentage of Al, Mg, Ca, Na, and K in the soil exchange complex (%) under each treatment in 2012, 2013 and 2014. HP: bio-waste based fertiliser with high pH, LP: bio-waste based fertiliser with low pH, T1, T2.5, T5, T10: bio-waste based fertilisers doses (1, 2.5, 5 and 10 Mg ha^{-1}) combined with mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O)), NF: no fertiliser, MIN: mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O) with lime (2.5 Mg CaCO3 ha^{-1}) (+lime) and without lime (-lime); Different letters indicate significant differences between treatments within the same year and treatments are no significantly different if no letters are shown.

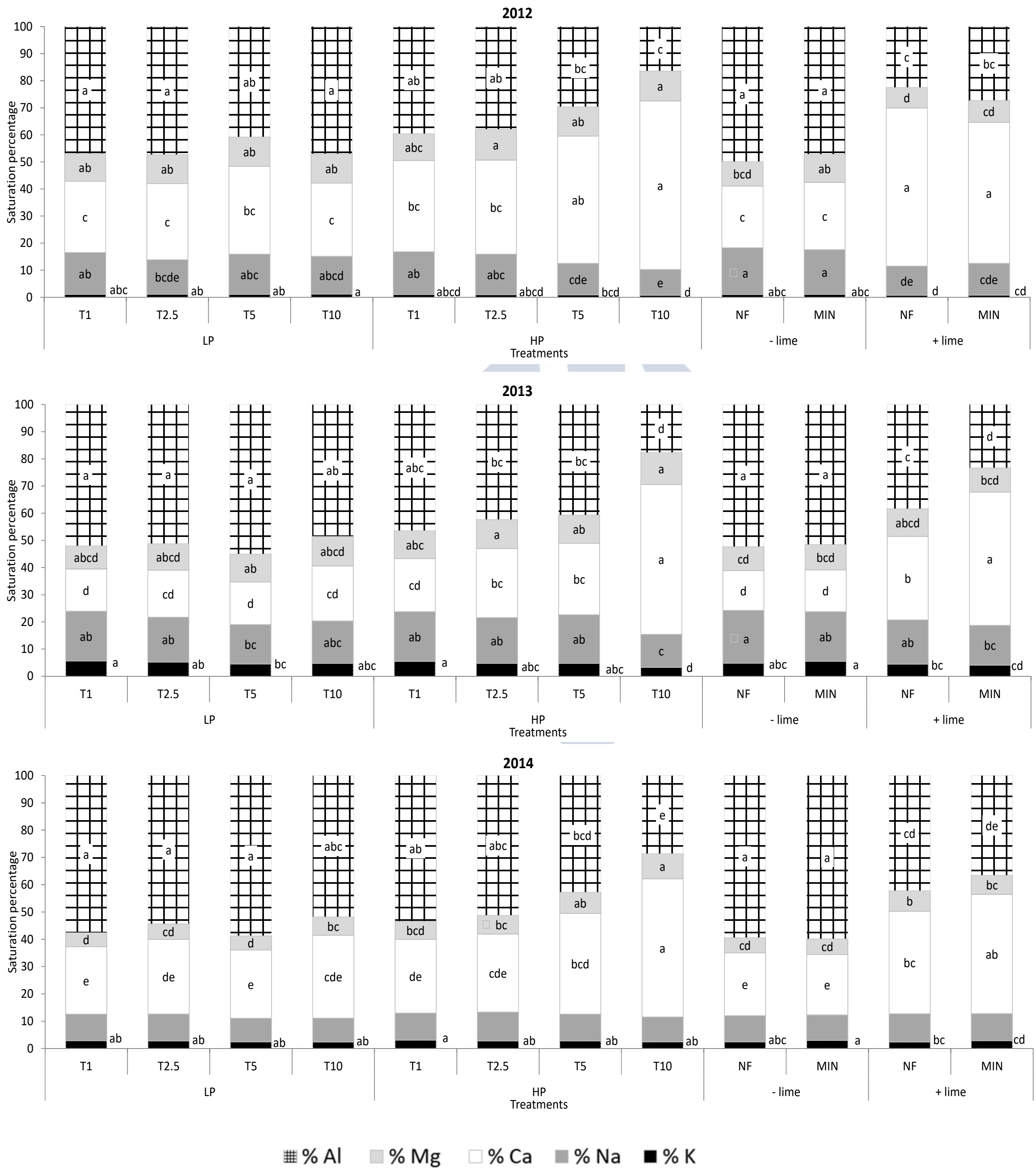


Figure 5. Pasture production under each treatment in 2012 and 2013. HP: bio-waste based fertiliser with high pH, LP: bio-waste based fertiliser with low pH, T1, T2.5, T5, T10: bio-waste based fertilisers doses (1, 2.5, 5 and 10 Mg ha^{-1}) combined with mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O)), NF: no fertiliser, MIN: mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O) with lime (2.5 Mg CaCO3 ha^{-1}) (+lime) and without lime (-lime). Different letters indicate significant differences between treatments within the same year. Bars indicate the standard error of the mean.

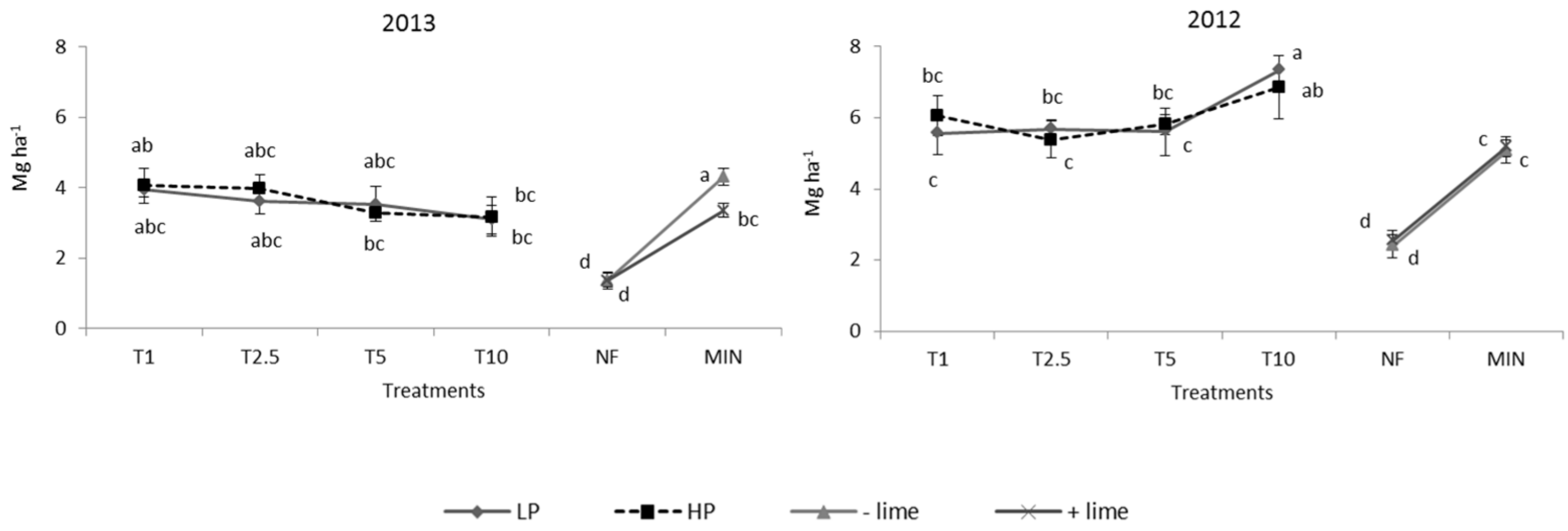


Figure 6. Evolution of the number of species under each treatment in 2012 and 2013. HP: bio-waste based fertiliser with high pH, LP: bio-waste based fertiliser with low pH, T1, T2.5, T5, T10: bio-waste based fertilisers doses (1, 2.5, 5 and 10 Mg ha^{-1}) combined with mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O)), NF: no fertiliser, MIN: mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O) with lime (2.5 Mg CaCO3 ha^{-1}) (+lime) and without lime (-lime).Different letters indicate significant differences between treatments within the same year. Bars indicate the standard error of the mean.

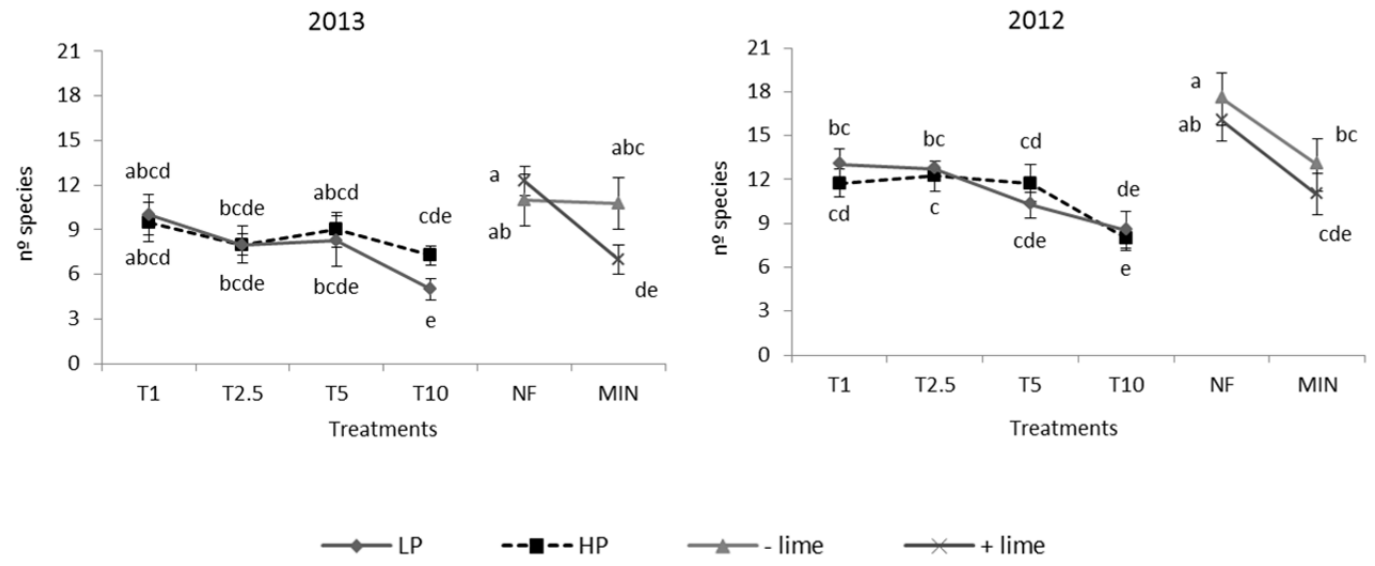


Figure 7. Abundance diagrams (relative proportion of each species based on dry weight) in 2012. HP: bio-waste based fertiliser with high pH, LP: bio-waste based fertiliser with low pH, T1, T2.5, T5, T10: bio-waste based fertilisers doses (1, 2.5, 5 and 10 Mg ha^{-1}) combined with mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O)), NF: no fertiliser, MIN: mineral fertiliser (500 kg ha^{-1} 8:24:16 (N:P2O5:K2O) with lime (2.5 Mg CaCO3 ha^{-1}) (+lime) and without lime (-lime). Species codes are shown in Table 4.

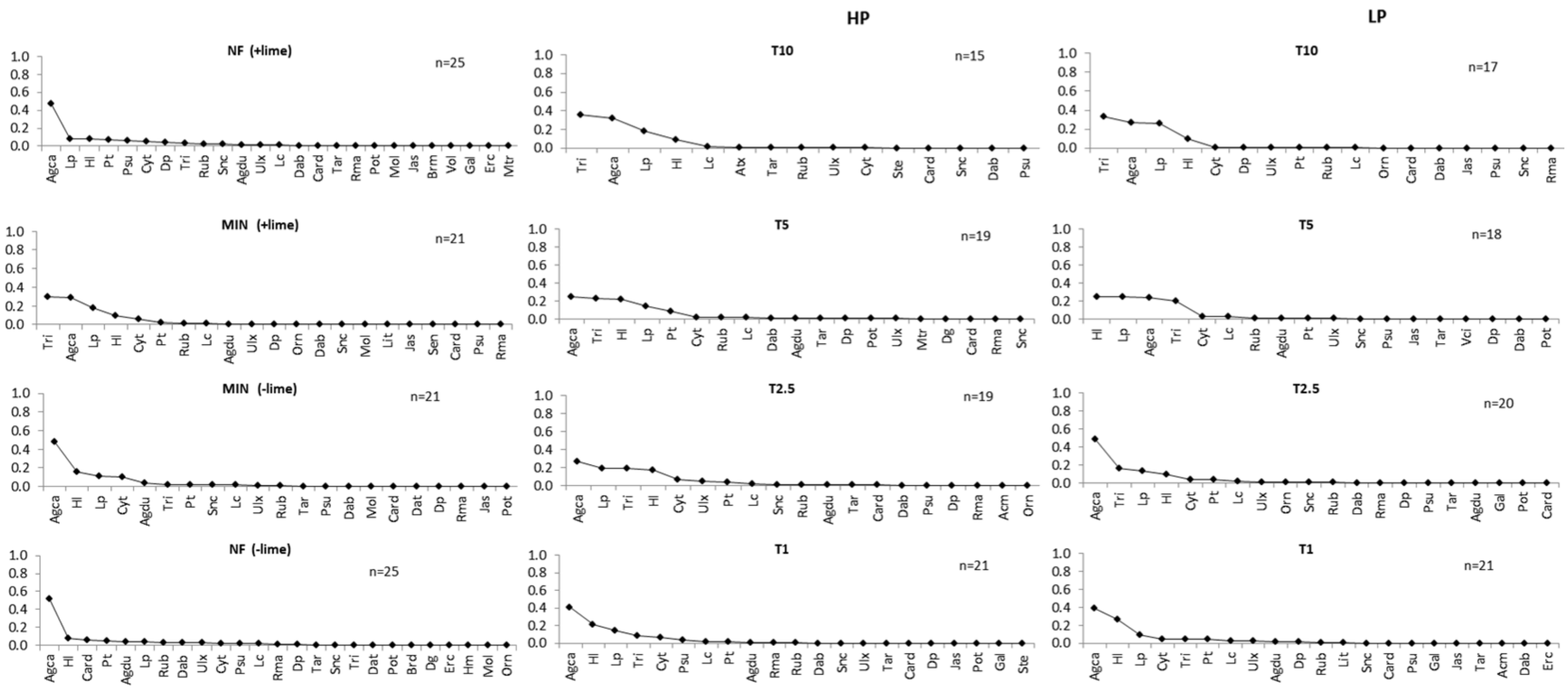
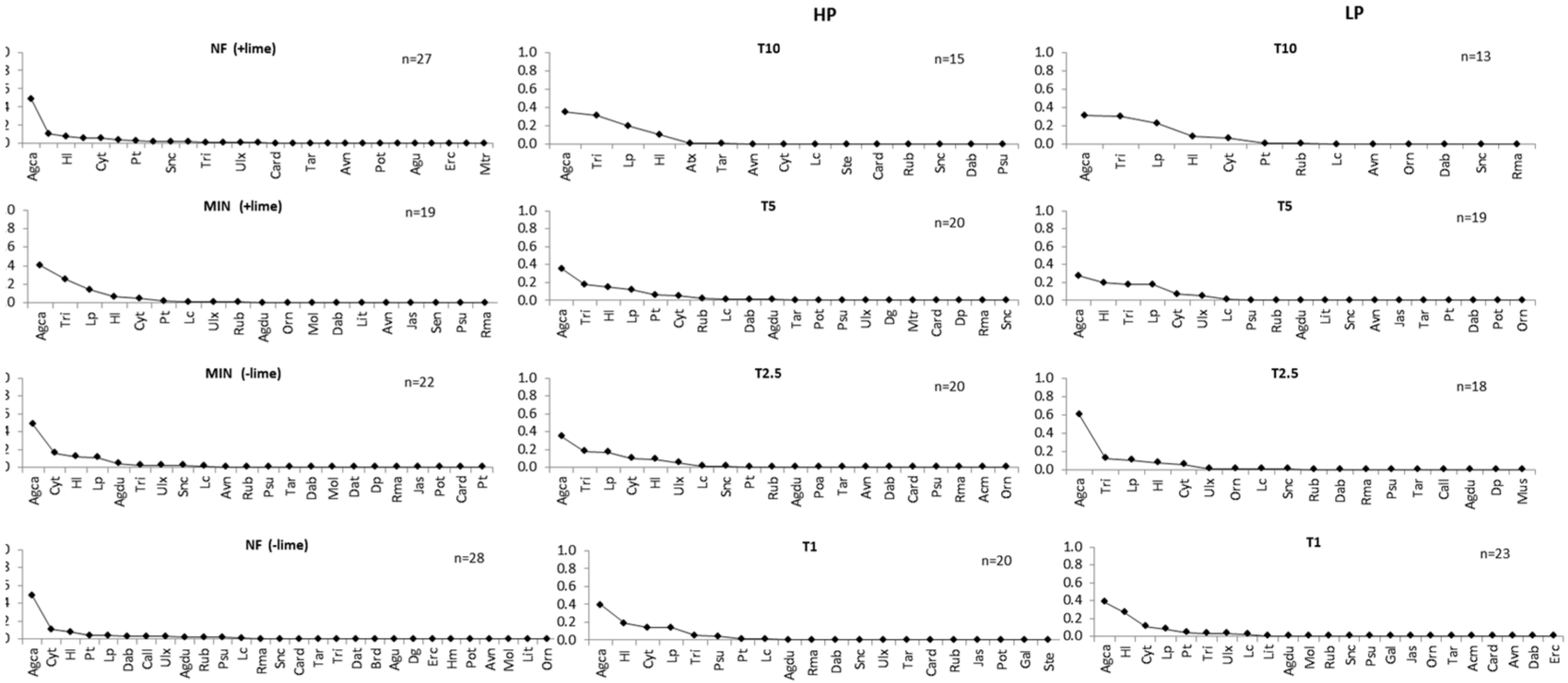


Figure 8. Abundance diagrams (relative proportion of each species based on dry weight) in 2013. HP: bio-waste based fertiliser with high pH, LP: bio-waste based fertiliser with low pH, T1, T2.5, T5, T10: bio-waste based fertilisers doses (1, 2.5, 5 and 10 Mg ha^{-1}) combined with mineral fertiliser (500 kg ha^{-1} 8:24:16 ($N:P_2O_5:K_2O$)), NF: no fertiliser, MIN: mineral fertiliser (500 kg ha^{-1} 8:24:16 ($N:P_2O_5:K_2O$) with lime (2.5 Mg $CaCO_3$ ha^{-1}) (+lime) and without lime (-lime). Species codes are shown in Table 4.



DISCUSIÓN

La discusión de la tesis se estructurará atendiendo a la secuenciación de los capítulos presentados previamente. La primera parte de la discusión se centra en la discusión sobre los principales resultados obtenidos en la evaluación de los suelos naturales agrarios de Galicia como posibles receptores de lodo. En la segunda parte, nos centraremos en el análisis de la aplicación de lodos en el suelo sobre la evolución de los niveles del metal pesado más limitante para el aporte de lodos en Galicia, el Zn, teniendo en cuenta los resultados de suelos del primer capítulo, los resultados medios de análisis de lodos desarrollados en toda España (Mosquera-Losada et al. 2010) y la normativa vigente. Finalmente, realizaremos un análisis del aporte de lodo estabilizado con cal sobre la producción y biodiversidad del pasto resultante de la transformación de zona de monte.

1. Empleo de lodos de depuradora en suelos agrarios de Galicia

A partir de la evaluación de los niveles de metales en los 2597 suelos de Galicia que nunca han recibido lodo en Galicia, se observa que los rangos de metales pesados del suelo encontrados estaban dentro de los descritos generalmente para Cd (Page et al., 1981) (0,01-3 mg kg^{-1}), Pb (Pais & Jones, 1997) (3-189 mg kg^{-1}) , Cr (Alloway, 1995) (0,3-10000 mg $kg^{-1)}$ y Hg (Pais & Jones, 1997) (0,01-1.8 mg kg^{-1}). Sin embargo, los elementos esenciales para plantas como Ni (Alloway, 1995) (1-100 mg kg^{-1}), Cu (Domínguez-Vivancos, 1997) (3-100 mg kg^{-1}) y Zn (Barber, 1995) (10-300 mg kg^{-1}) tienen un rango más amplio que los valores dados en la literatura para suelos. La asociación de altos niveles de metales pesados con suelos derivados de rocas ultrabásicas y rocas básicas del suelo (Ross, 1994; Díez-Lázaro et al., 2002; Candeias et al., 2011) podría explicar el rango más amplio de Ni y Zn como demuestra el análisis factorial llevado a cabo. Por otra parte, todos los rangos de metales pesados encontrados en estos suelos agrícolas eran más amplios que los detectados en el estudio de suelos sistemático desarrollado por Macías-Vázquez y Calvo de Anta (2009) para todos los tipos de suelos gallegos (no sólo agrícolas). Sin embargo, los valores medios de los metales pesados encontrados fueron inferiores a la media para todos los tipos de suelos gallegos (Macías-Vázquez & Calvo de Anta, 2009). Por otra parte, de los suelos agrícolas evaluados en nuestro estudio, el 90 % tiene valores de Cd (0,10 frente a 0,11) y Pb (27,12 frente a 29,32) por debajo de la media gallega en la que la proporción de suelos procedentes de rocas ultrabásicas fue mayor. Las diferencias entre los suelos que cumplen o no la regulación RD 1310/1990 fueron mayores en Ni que en Cu, Cr, Zn y Pb, en este orden. Los niveles de Ni en los suelos derivados de granito, el esquisto la pizarra y sedimentos suelen ser superiores a 30 mg kg^{-1}, ya que son 23,7, 43,2, 40,8 y 29,2, respectivamente, siendo el valor medio de las rocas ultrabásicas alrededor de 2155 mg kg^{-1} (Macías -Vázquez & Calvo de Anta, 2009). Esto explica por qué la mayoría de las muestras que no cumplen RD 1310/90 se vinculan a concentración de Ni en los suelos naturales gallegos, a los que previamente no se había añadido lodo. El resto de las muestras que no cumplen con los requisitos de Cu, Cr, Zn y Pb también se pueden asociar con áreas de rocas ultrabásicas y básicas como material de origen en los lugares donde se encuentran.

Nuestros resultados mostraron que más del 90% de los suelos agrícolas gallegos son adecuados para recibir lodos de depuradora según la normativa vigente, lo que significa que algunos de los suelos podrían aumentar sus contenidos en metales pesados por encima de los niveles basales para los suelos gallegos dados por Macías-Vázquez y Calvo de Anta (2009). Sin embargo, el hecho de que todos los valores medios y medianos de los suelos agrícolas del estudio desarrollado en suelos agrícolas de Galicia estuvieran por debajo de los niveles de referencia de metales pesados de todos los tipos de suelos gallegos, indica que la mayoría de los suelos gallegos son adecuados para recibir fertilización con lodos de depuradora sin aumento significativo de los niveles habituales de metales pesados en estos suelos, y siempre y cuando se efectúe una aplicación en dosis adecuada de lodos de depuradora de elevada calidad (bajos niveles de metales pesados) para satisfacer las necesidades de los cultivos.

Considerando las características químicas medias de los lodos de depuradora en España (Mosquera-Losada et al., 2010) estabilizado mediante digestión anaerobia, podemos estimar la cantidad de lodos de depuradora que se debe aplicar en un suelo con una tasa de mineralización de nitrógeno dada para cubrir las necesidades del cultivo (EPA, 1994). Una vez conocida la dosis y las concentraciones de metales pesados de los lodos de depuradora, podemos obtener la cantidad de metal pesado que incorporamos por aplicación y el número mínimo de aplicaciones que se pueden realizar para alcanzar las concentraciones máximas en suelo de metales pesados permitidas por el Real Decreto 1310/90 en suelos ácidos, suponiendo que no hay lixiviación o extracción de cultivos de metales pesados procedentes de suelos (McGrath, 1987). Las aplicaciones de lodos de depuradora anaerobios en dosis de 200 kg de N ha^{-1} total (EPA, 1994) en un suelo medio gallego, implican un aumento total de 1,26, 0,37, 0,05, 0,01, 0,16, 0,001 y 0,03 mg de Zn, Cu, Ni, Cd, Pb, Hg y Cr por kg^{-1} de suelo y aplicación, respectivamente (densidad normal del suelo: 1,1 Mg m^3 y profundidad de 0,25 m (RD 1310/90)). Estas cifras permiten un número mínimo de aplicaciones de Zn, Cu, Ni, Cd, Pb, Hg y Cr en un suelo medio de 82, 89, 304, 94, 253, 491 y 2474 años, respectivamente. Por tanto, teniendo en cuenta los valores medios de concentración en metales pesados de los lodos y de los suelos considerados en el presente estudio, los nutrientes esenciales Zn, Ni y Cu son los más limitantes en la aplicación de lodos de depuradora, como se ha encontrado en suelos evaluados después de realizar aplicaciones de lodos de depuradora a largo plazo (McGrath, 1987). Entre todos los parámetros del suelo que podrían modificar el destino final de los metales pesados del suelo, sólo el pH del suelo está actualmente incluido en la mayoría de las regulaciones para el uso de lodos de depuradora en la agricultura. El pH del suelo afecta directamente la disponibilidad de metales pesados (Parat et al., 2005, Lu et al., 2012) y, por tanto, su destino: lixiviación o extracción por parte del cultivo. Es sabido que la mayoría de los metales pesados precipitan en suelos neutros y básicos como se encuentran en experimentos a largo plazo (McGrath, 1987), y sólo el movimiento del suelo como el arado podría explicar el agotamiento de los metales pesados (McGrath, 1987). Por el contrario, en suelos ácidos y suelos con una reducción gradual de la materia orgánica, un efecto llamado "sludge time bomb" (bomba de tiempo de lodo) podría ser de gran relevancia ya que la mineralización de materia orgánica podría liberar formas más solubles de metales pesados que podrían ser lixiviados o extraídos posteriormente por los cultivos. Esto podría explicar, por ejemplo, por qué las poblaciones totales de bacterias del suelo mejoran inicialmente después de la incorporación de nutrientes con las aplicaciones de lodos de depuradora y luego se agotan cuando aumenta la disponibilidad de metales pesados en los suelos (Guiller et al., 2009).

El concepto de "sludge time bomb ", que podría estar más asociado a los suelos ácidos, hace importante proponer criterios para las aplicaciones de lodos en el suelo, que aseguren que los valores promedio de metales pesados para cada tipo de suelo no se superen. Tener los valores medios del suelo para cada tipo de material de roca madre como criterio para aplicar lodos de depuradora probablemente asegura que las poblaciones microbianas y de vegetales, puedan sobrevivir en los entornos ya existentes, promoviendo así la sostenibilidad y preservando la biodiversidad alpha y beta entre diferentes tipos de condiciones del suelo. Además, la preservación de los valores medios del suelo de los metales pesados para cada tipo de material de roca madre, también será adecuada para la sostenibilidad de la producción agrícola de alimentos para consumo humano o animal, ya que estos suelos se emplean habitualmente en la producción de cultivos.

La normativa vigente en la UE y en España se basa en un rango de pH amplio. La mayoría de los suelos gallegos tienen un pH ácido, como ocurre con los suelos agrarios evaluados en este estudio (sólo el 1,5% de las muestras de suelo tienen un pH superior a 7). El borrador de reglamento EUDWD (EC, 2000) no establece los niveles de metales pesados por debajo de los cuales el lodo podría aplicarse si el pH del suelo es inferior a 5, lo que afecta directamente al 41% de los suelos gallegos que podrían ser receptores de este residuo. Aunque el pH ácido del suelo es un factor clave para aumentar la solubilidad de los metales pesados en el suelo (Parat et al., 2005) y por lo tanto su potencial de ser lixiviado y extraído por los cultivos, tiene aspectos importantes a tener en cuenta a la hora de realizar aplicaciones de lodos de depuradora en suelos ácidos. Los suelos naturales gallegos generalmente están por debajo del pH 5 cuando no se aplica cal, principalmente debido al régimen de lluvias y la extracción del cultivo. Las recomendaciones para la aplicación de la cal cada cuatro años en Galicia suelen basarse en la reducción del porcentaje de saturación de Al por debajo del 20%, que puede obtenerse con frecuencia si el pH del suelo es inferior a 5,5. Si se aumenta artificialmente el pH del suelo con aplicaciones de cal, se permiten aplicaciones de lodos de depuradora y, por lo tanto, el proceso de "sludge time bomb " podría iniciarse fácilmente una vez finalizado el efecto del encalado (normalmente de 4 años). De los suelos analizados en el presente estudio, 41% y 88% tenían un pH inferior a 5 y 6, respectivamente. Esto significa que si se usó lodo de depuradora porque el pH del suelo es superior a 5 y no se aplica cal durante un largo período de tiempo, entonces el pH del suelo podría volver a reducirse fácilmente. Por otro lado, aunque la mayoría de los metales pesados están más disponibles a pH bajos del suelo, algunos como el Cu podrían estar menos disponibles por debajo de 5 que por encima de este valor (Porta, 2010). Por lo tanto, puede ser mejor tener en cuenta los niveles de referencia existentes de metales pesados en los suelos y controlar la dosis máxima que se permite aplicar mediante el establecimiento de un porcentaje de los niveles de referencia. Esto permitirá la aplicación de cantidades más específicas de lodos de depuradora, dependiendo del tipo de suelo y de su material parental, evitando incrementos significativos de los metales pesados del suelo de manera controlada, lo que al mismo tiempo protegería al ecosistema y probablemente reduciría los impactos negativos a largo plazo de las aplicaciones de lodos de depuradora en el suelo y en los microorganismos fijadores de nitrógeno (Guiller et al., 2009). Los niveles medios de referencia para cada tipo de suelo permitirán probablemente la aplicación de lodos de depuradora en más del 44,37% (473/1066) de suelos con pH inferior a 5, ya que todos los metales pesados medios están por debajo de los umbrales más bajos establecidos por el borrador de reglamento EUDWD (EC, 2000). Por otra parte, la reducción en la aplicación de lodos, recomendada por la EUDWD, ha evitado la necesidad de aprobar el borrador EUDWD, publicado hace más de diez años. Además, es importante recomendar otras prácticas, como el uso de lodos de buena calidad, la incorporación de lodos de depuradora en suelos mediante arado para evitar un gradiente de lodos de depuradora y prohibir el pastoreo directo después de las aplicaciones, ya que los animales consumen grandes cantidades de suelo (entre 182 y 803 kg al año en vacas lecheras (Herlin & Andersson, 1996)) y por lo tanto de metales pesados presentes en los lodos aplicados. También debe reducirse la frecuencia de aplicación en el mismo terreno.

2. Análisis del aporte de Zn con lodos de depuradora urbana en suelos gallegos

Una vez evaluados el potencial de uso de los lodos (Mosquera-Losada et al. 2016) en los suelos en relación a los niveles de metales pesados existentes como nivel base, el siguiente objetivo planteado fue analizar los límites actuales y previsibles de la aplicación de lodos de depuradora, considerando la legislación actual y futura del metal pesado más abundante en el lodo de aguas de depuradora (Zn) considerando los niveles existentes en los suelos ácidos en Galicia analizados, y el uso de 3 tipos de lodos estabilizados, que son los más extensamente empleados): digestión anaerobia (anaerobio), compostaje (compost) y secado térmico-pellet- (pellet).

De tal forma que se garantice la salud microbiana del suelo y unos niveles adecuados del Zn en el pasto y en los cultivos, ya que la calidad de los lodos de depuradora, entendida como la concentración de metales pesados, varía entre los tipos de lodos y modifica el uso potencial de los lodos de depuradora en la agricultura (Mosquera-Losada et al., 2010).

A pesar de que la calidad de los lodos de depuradora utilizados en este estudio es adecuada (Mosquera-Losada et al., 2010) y con una concentración de metales pesados que se encuentra muy por debajo de los indicados por la normativa vigente (JRC, 2012), existen serias restricciones para su uso como fertilizante si se consideran otros aspectos regulados como la cantidad máxima de Zn que puede ser aplicado en el suelo como media durante un período de diez años, o la concentración de Zn en el suelo. La normativa actual permitiría en la mayoría de los casos aplicaciones en suelos durante un período de diez años de pellet y anaerobio. Sin embargo, las entradas de Zn con compost están seriamente limitadas por la norma que limita la cantidad máxima de metal a aportar en un período de 10 años del Real Decreto actual, como consecuencia de las grandes cantidades de compost necesarias para disponer de suficiente nitrógeno para satisfacer las necesidades de los cultivos, debido a la baja concentración de nitrógeno que tiene y a la reducida capacidad de mineralización (Yongjie y Yangsheng, 2005). Otros autores como Mosquera-Losada et al. (2010) encontraron un resultado similar con la evaluación de la calidad de los lodos de depuradora, lo que se justifica por el menor grado de nitrógeno (N) disponible a través de la mineralización cuando se aplica compost en comparación con anaerobio o pellet (Mosquera-Losada et al., 2017). Esto hace aconsejable utilizar el compost como una enmienda orgánica del suelo (en lugar de un fertilizante) ya que la persistencia de materia orgánica en el suelo será más larga que con anerobio y pellet debido a la alta proporción de lignina procedente del resto de las ramas usadas para hacer compost con el lodo (Mosquera-Losada et al., 2017). En el caso de tener en cuenta el borrador de reglamente EUDWD (EC, 2000), sólo se permitirían dosis bajas de aplicación de N con anaerobio durante 10 años consecutivos, siendo por lo tanto más restrictivo que la regulación actual y limita seriamente el uso de cualquier tipo de lodos de depuradora como fertilizante. Se pueden contemplar dos soluciones para cumplir la regla de los 10 años en EUDWD (EC, 2000), una sería mejorar la calidad de los lodos de depuradora y la segunda aplicar lodos de depuradora con una frecuencia inferior a un año y combinarla con la aplicación de fertilizantes inorgánicos en los años intermedios para satisfacer la necesidad de N de los cultivos. También podría ser posible una combinación anual de N inorgánico con el N orgánico proveniente de los lodos de depuradora, aumentando la disponibilidad del N gracias a la reducción de la relación C/N cuando ambos tipos de fertilizantes se combinan entre sí.

Todas las muestras de los más de dos mil quinientos suelos analizados cumplen la normativa vigente en relación a los niveles de Zinc, Directiva UE 86/278 / CEE (UE, 1986) y española RD 1310/1900 (BOE, 1991) (Zn: 150-450 mg kg^{-1}). Sin embargo, todos los suelos con un pH inferior a 5 no podrán recibir lodos de depuradora siguiendo el borrador EUDWD (EC, 2000) y sólo el 21,3% de los suelos pueden considerarse adecuados para recibir lodos de depuradora si se tiene en cuenta, para aquellos suelos con un pH inferior a 5, el límite establecido por EUDWD (EC, 2000) para suelos con pH entre 5 y 6. Cuando se considera el escenario menos (S1,“Moderate changes”) y más restrictivo (S2,“More significant changes”) propuesto por el JRC (EC, 2008), sólo el 1,9%, y el 69,7% respectivamente no recibirían lodos de depuradora cuando el pH del suelo es inferior a 5 (considerando los valores límite para el pH entre 5 y 6). Con respecto a los límites de Zn para el pH del suelo entre 5 y 6, se obtuvo un resultado similar (24,9, 2,3 y 79,8% para EUDWD (100 kg^{-1}), S1 (100 mg kg^{-1}) y S2 (20 mg kg^{-1}) respectivamente, no podrían recibir los lodos de depuradora). La mediana de la concentración de Zn en suelo encontrada en el segundo capítulo de la tesis (pH <5, 39,68 mg kg^{-1}, 5≤pH <6, 44,00 mg kg^{-1}) estaba muy por debajo de los suelos naturales mundiales (Barber (1995): 10-300 mg kg^{-1}) y Kabata-Pendias y Pendias (2001): 10-105mg kg^{-1})), en los suelos agrícolas y de pastizales con pH inferior a 7 en España (58,42 mg kg^{-1}) (Rodríguez Martín et al) y en el 98% de los suelos naturales gallegos (NW de España) (Macías-Vázquez y Calvo et al., 2009: 100 mg kg^{-1}). Los suelos gallegos agrarios actuales son cultivados con niveles de Zn por encima del límite propuesto en S2 (Mosquera-Losada et al., 2017), mostrando en varios casos un déficit de Zn en el pasto, que se necesita y se considera micronutriente para alimentar a los animales. Este déficit puede ser explicado por la gran cantidad de materia orgánica (SOM) en suelos gallegos (alcanzando valores del 20%) como mencionó Sánchez-Rodríguez et al. (2002), ya que la SOM adsorbe Zn (Alloway 1995, Kabata-Pendias y Pendias, 2001, McBride et al., 2004) y no está disponible para los cultivos (Mosquera-Losada et al., 2009). El alto contenido de SOM en Galicia puede explicarse por la alta acidez de los suelos que impide la mineralización de SOM debido a la baja presencia de bacterias en el suelo para mineralizarla. Por tanto se considera que la SOM es un aspecto relevante a tener en cuenta en la futura normativa europea de lodos de depuradora en Europa, ya que el 35,6% de los pastizales y el 25,8% de los suelos europeos son ácidos (Clea et al., 2014).

La evaluación de los límites establecidos en la normativa actual y futura si tenemos en cuenta los límites para Zn en el suelo y los diferentes tipos de lodo (compost, anaerobio y pellet), con y sin extracción de cultivos de Zn, muestra que el compost es el que tiene más limitado su uso en agricultura, después el pellet y finalmente el anaerobio. Considerando diferentes dosis (50, 100, 150 y 200 kg N disponible ha^{-1}) y una aplicación anual de lodo de depuradora s con la legislación actual, se alcanza el límite de Zn en suelo antes con el compost (6-21 años) antes que con el pellet (10-41 años) o el lodo anaerobio (16-68 años). Si consideramos la reglamentación de EUDWD (EC, 2000) para suelos ácidos (pH <6), el compost podría ser utilizado durante 4 años, pellet durante 8 años y anaerobio durante 13 años. El compost es el que satisface los requerimientos de nutrientes del cultivo (Mosquera-Losada et al., 2017) por el incremento de Zn aplicado en el suelo, por lo que se recomienda el empleo del compost con reducidas dosis de aplicación (Yongjie y Yangsheng, 2005) justificada como enmienda más que como fertilizante para mejorar la estructura y la capacidad de intercambio catiónico del suelo. Por lo tanto, el EUDWD (CE, 2000) restringe la reutilización de lodos de depuradora en la agricultura como fertilizante más que la Directiva 86/278/CEE (UE, 1986) traspuesta en el RD 1310/90 y eso, a pesar de tratar de fomentar el uso de lodos de depuradora como parte de paquete sobre Economía Circular (CE, 2015). Sin embargo, S1 permite el uso de lodos de depuradora durante más años que EUDWD en suelos ácidos (pH<6). En el marco del escenario S2, no está permitido utilizar los lodos de depuradora en agricultura, a pesar de la alta calidad de los suelos gallegos y el bajo contenido medio en los lodos de depuradora españoles en Zn, en comparación con los suelos y lodos de otras áreas.

Por otra parte, el uso de lodos de depuradora ha mostrado un claro beneficio para los cultivos no alimentarios (árboles forestales) y pastizales cuando se aplican en el primer año o durante varios años consecutivos con una misma dosis total (Mosquera-Losada et al., 2009, 2011a, 2012b, 2017) en experimentos a largo plazo realizados en suelos ácidos. Los contenidos de Zn del suelo no se incrementaron peligrosamente incluso cuando se aplicaron mayores cantidades de N (120 N). Por otra parte, hay suelos naturales en Galicia y otras zonas europeas con altos niveles de Zn que se utilizan actualmente para la producción de alimentos directamente consumidos por los seres humanos. Estos suelos tienen poblaciones microbianas específicas del suelo que deben ser preservadas. Por lo tanto, una buena propuesta para regular Zn o cualquier aporte de metales pesados de los lodos de depuradora en suelos no debería exceder los niveles medios de los suelos asociados a materiales parentales específicos (Mosquera-Losada et al., 2017), así como aplicaciones de compost como enmendante en años no consecutivos que podrían ayudar a enriquecer la SOM, y mejorar los beneficios que esta práctica proporciona como mejoradora de la fertilidad del suelo, mantener los niveles naturales de Zn y satisfacer las necesidades de los animales.

3. Uso del lodo en la transformación de matorral a pasto

El aporte de enmienda orgánica, elaborada con lodo estabilizado mediante cal, en la transformación de monte en pastos provocó un aumento del pH del suelo, CIC (Capacidad de Intercambio catiónico) y porcentajes de saturación de K y Na dos años después de su aplicación, siendo este aumento muy leve en el caso del pH del suelo. Autores como Diacono et al. (2010) han demostrado en sus estudios que la adición regular de enmiendas orgánicas, basadas en residuos orgánicos biodegradables, al suelo aumenta la fertilidad física del mismo, principalmente debido a la mejora de la estabilidad de los agregados del suelo. La aplicación al suelo de enmiendas orgánicas, también puede aumentar la disponibilidad de K, P extraíble, carbono orgánico y el contenido de N orgánico del suelo, sin elevar la lixiviación de nitratos a las aguas subterráneas (Diacono et al., 2010). Además, la mejora de estas variables del suelo a principios de segundo año (2013), en comparación con el primero tras el aporte de la enmienda orgánica (2012), podría explicarse también por un aumento en la tasa de mineralización de la materia orgánica del suelo debido a (i) la aplicación de fertilizantes minerales en todas las parcelas y (ii) al aumento de la humedad del suelo derivada por una mayor precipitación en 2012 (1179,9 mm) que en 2011 (966,4 mm). La humedad del suelo es uno de los principales factores que controlan la mineralización y este factor es clave en la regulación del ciclo de los nutrientes del suelo (EPA, 1994). Por lo tanto, el aumento de la tasa de mineralización podría haber favorecido la liberación de nutrientes en el suelo, lo que mejoró la CIC. Resultados similares fueron descritos también por Ferreiro-Domínguez et al. (2014) en un sistema silvopastoral establecido en la misma región y fertilizado con diferentes dosis de lodos de depuradora digerido anaeróbicamente (50 y 100 kg de N ha^{-1} total) combinado con encalado. Además, los mayores porcentajes de saturación de K y Na a principios de 2013, que en 2012, se explican por la adición de K inorgánico a partir de fertilizantes minerales y la facilidad de liberación de Na de la enmienda orgánica. El aumento de K y Na probablemente explique por qué el porcentaje de saturación de Ca disminuyó a principios de 2013, en comparación con 2012, debido a que este catión se caracteriza por su fuerte antagonismo con el K (Barber, 1995, Mosquera-Losada et al., 2012) y el Na (Kopittke, 2012). Sin embargo, a comienzos de 2014, el pH del suelo, la CIC y los porcentajes de saturación de K, Mg y Na disminuyeron en comparación con febrero de 2013, lo que podría explicarse por diversos factores. En primer lugar, la alta precipitación registrada durante 2013 (1744,1 mm), principalmente en los últimos meses del año, lo cual podría haber favorecido la lixiviación de los cationes a través del perfil del suelo, siendo el orden de solubilidad de los cationes CIC, Na+ >K+ >Mg+ >Ca2+ >Al3+ (Calvo et al., 1987, Álvarez et al., 1992). En segundo lugar, debido a que las enmiendas orgánicas sólo se aplicaron al inicio del estudio, sugiere que los pastizales podrían haber extraído los cationes (Adams et al., 2001). La extracción de los cationes del suelo por el pasto también podría explicar el bajo porcentaje de saturación de Ca al final del experimento, ya que el pasto tiene niveles más altos de Ca que K y Na, y el consiguiente incremento del porcentaje de saturación de Al a lo largo del tiempo, lo cual se observó principalmente en los tratamiento sin encalado y con enmiendas orgánicas. Estos resultados fueron previamente descritos por otros autores como Ferreiro-Domínguez et al. (2011) y Mosquera-Losada et al. (2012) en sistemas silvopastorales establecidos bajo Pinus radiata D. Don en suelos ácidos de Galicia.

Por otra parte, la mejora de la fertilidad del suelo causada por la aplicación de las enmiendas orgánicas probablemente aumentó la producción de pastos en 2012. Sin embargo, el efecto residual en la mejora de la fertilidad del suelo encontrado en febrero de 2013, no afectó a la producción de 2013. Los cambios en la fertilidad del suelo probablemente también expliquen el mayor número de especies en 2013 (40 especies) que en 2012 (36 especies). Las condiciones edáficas menos restrictivas podrían haber favorecido el establecimiento de especies de gramíneas como *Agrostis curtisii* Kerguelen, *Avenula sulcata* (Gay ex Boiss) Dumort y *Poa annua* L. en 2013. Además, la mayor precipitación en 2013 que en los otros años del experimento probablemente promovió el establecimiento de musgo en ese año. Estos resultados demuestran que la composición botánica del pasto fue más sensible a la variación de la fertilidad del suelo causada por la aplicación de enmiendas orgánicas que la propia producción de pasto. Por lo tanto, la composición botánica podría ser considerada como un buen bioindicador de los cambios en la fertilidad del suelo cuando se utilizan las enmiendas orgánicas en la agricultura. El efecto positivo de la enmienda orgánica sobre el pasto y su composición fue observado previamente por varios autores en la misma región (Mosquera-Losada et al., 2009; Ferreiro-Domínguez et al., 2011). Sin embargo, el número medio de especies por tratamiento y año disminuyó con el tiempo, lo que podría explicarse por una reordenación de las especies debido al cambio de dominancia.

La comparación del efecto de la aplicación de cal y de las diferentes dosis de las enmiendas orgánicas en las variables del suelo mostró un efecto positivo del encalado y las dosis altas (T10:10 Mg ha^{-1}) de la enmienda orgánica con mayor pH (HP) en el pH del suelo, CIC y porcentaje de saturación de Ca y de Mg. El efecto positivo de la cal sobre estas variables del suelo podría deberse al aumento del Ca intercambiable (Slattery et al., 1995), que tiende a mejorar las propiedades físicas y químicas del suelo y la actividad microbiana (Baley, 1995; Álvarez et al. Al., 1992), causando mineralización orgánica y por lo tanto, la liberación de nutrientes (Wheeler, 1998). Resultados similares también se encontraron en experimentos realizados en la misma región, donde se evaluó el efecto de la cal sobre la producción de pastizales y el crecimiento de Pinus radiata D. Don (Ferreiro-Domínguez et al., 2014). En el caso de las enmiendas orgánicas, la mejora del pH del suelo, la CIC y los porcentajes de saturación de Ca y Mg cuando se aplicaron altas dosis de HP (T10) en comparación con las otras dosis de enmienda orgánica con pH alto (HP) y con pH abajo (LP), podría explicarse por el pH más alto del HP (13,19) que del LP (6,96). Por otra parte, la dosis alta de HP (T10) implicó mayores aportes de Ca y Mg en el suelo (2774 kg Ca ha^{-1} y 200 kg Mg ha^{-1}) que las otras dosis de HP y las otras dosis de LP (T10: 549 kg Ca ha^{-1} y 41 kg Mg ha^{-1}). Además, el aumento del porcentaje de saturación de Ca, después de la aplicación de cal y altas dosis de HP, disminuyó los porcentajes de saturación de Al, K, Mg y Na debido al antagonismo entre estos cationes y el Ca (Keltjens y Tan, 1993, Smith, 1996, Prasad y Power, 1997, Fageria, 2001) y la energía más fuerte de adsorción de Ca que Mg, K o Na (Foth, 1990). Finalmente, la fertilidad del suelo fue generalmente similar en las parcelas que recibieron dosis altas de HP en comparación con las parcelas en las que se llevaron a cabo las prácticas de manejo convencionales de la región, como es el encalado o la fertilización mineral combinada con cal. Es importante tener en cuenta que la enmienda orgánica con pH más alto HP, al igual que sucede con otros lodos de depuradora estabilizados con cal, tiene una equivalencia y eficacia inferior a la cal (CaCO3) (Sloan y Basta, 1995; Guo et al., 2012), y por esta razón es necesario aplicar una dosis más alta de HP que de cal, para aumentar el pH del suelo, la CIC y el porcentaje de saturación de Ca. Sin embargo, con la enmienda orgánica se aplica una mayor cantidad de materia orgánica y nutrientes que con la cal.

En este experimento se observó un claro efecto de los tratamientos establecidos sobre la producción y la biodiversidad del pasto. En general, la fertilización mineral aumentó la producción de pasto en comparación con el tratamiento NF, en el que las propiedades químicas del suelo siguen siendo muy pobres (baja CIC y alto porcentaje de saturación del Al). El efecto positivo de la fertilización mineral en la producción de pasto fue descrito previamente por numerosos autores (López-Díaz et al., 2007, Courtney y Mullen, 2008, Mosquera-Losada, et al., 2012, 2016) y podría explicarse por los insumos de N inorgánico al suelo, lo que reduce la relación C/N aumentando la mineralización de la materia orgánica del suelo, y por lo tanto la disponibilidad de cationes para el pasto (Whitehead, 1995). Por otra parte, en el primer año del estudio, la producción de pasto fue mayor cuando se aplicó la dosis alta (T10) de LP al suelo en comparación con las dosis más bajas de LP y HP, probablemente debido a que la dosis alta de LP aporto más N (101 kg N ha^{-1}) que las otras dosis de LP y HP (T5: 24 kg N ha^{-1}), siendo N el principal nutriente limitante para el crecimiento de las plantas (Burgos et al. , 2016, Obriot et al., 2016), y a que probablemente la mayor dosis de HP causó una mayor liberación de nutrientes del suelo a medida que se activaba la mineralización. Además, en general, en ambos años del experimento, la producción de pasto asociada a las dosis baja e intermedia (T1, T2.5 y T5) de las enmiendas orgánicas fue similar a la producción de pasto obtenida en el tratamiento mineral, lo cual es indicativo de la liberación de nutrientes de la materia orgánica del suelo en la región donde se realizó este experimento (López-Díaz et al., 2007; Mosquera-Losada et al., 2012). Otros autores como Herencia et al (2007) también encontraron en sus estudios que el uso de lodo de depuradora estabilizado mejora la fertilidad del suelo, y produce rendimientos y composición de nutrientes en los cultivos similares a la fertilización mineral. En cualquier caso, es importante tener en cuenta que cuando se aplicaron las dosis altas de las enmiendas orgánicas a base de lodos de depuradora estabilizados con cal (HP y LP) al suelo, la producción de pasto obtenida en este estudio en 2012 estuvo dentro del rango de producción de pastos encontrado en más de 80% de las parcelas de Galicia (6,25-18,75Mg ha^{-1}) MAPAMA (2013).

Sin embargo, la producción de pasto fue menor que el rango de producción de pastos establecido por MAPAMA (2013), cuando se aplicaron las otras dosis de las enmiendas orgánicas, y los tratamientos NF y MIN en 2012, y en todos los tratamientos en 2013. Estos Los resultados indican que, en primer lugar, hay un efecto positivo de la aplicación de altas dosis de estas enmiendas orgánicas en la producción de pastos en comparación con las prácticas de manejo convencional llevadas a cabo en la zona (fertilización mineral) y, en segundo lugar, la producción de pasto disminuye con el tiempo, por lo que es aconsejable su aplicación periódica. Por otra parte, los mayores aportes de N al suelo asociados a los tratamientos con mineral y con las altas dosis de las enmiendas orgánicas, probablemente disminuyeron el número de especies en comparación con los tratamientos con dosis bajas de las enmiendas orgánicas y de no fertilización (NF). Otros autores como Willems et al. Hyvonen y Salonen (2002) o Dise y Stevens (2005) también encontraron en sus estudios que los aportes de nutrientes al suelo desfavorecen la biodiversidad, aunque este efecto depende de varios factores, como las condiciones iniciales del suelo o la capacidad de adaptación de las especies a perturbaciones en un determinado contexto edafoclimático. Además del efecto de las dosis de las enmiendas orgánicas estudiadas sobre el número total de especies, también alteró la distribución de las especies con respecto a su proporción relativa. En general, *Agrostis capillaris* L. fue la especie más abundante en todos los tratamientos del estudio en 2012 y 2013, con la excepción en 2012 de los tratamientos con la dosis más alta (T10) de las enmiendas orgánicas (LP y HP), en los que *Agrostis capillaris* L. comparte dominancia con *Trifolium repens* L. El cambio en la proporción de *Trifolium repens* L. debido a la aplicación de altas dosis de HP y LP, puede explicarse por la mejora de la fertilidad del suelo asociada a estos tratamientos, ya que esta especie requiere más fertilidad en suelo que otras especies espontáneas como *Agrostis capillaris* L. (Grime et al., 2007), y es muy sensible a crecer en suelos con un alto nivel de saturación Al. Otros autores como Mosquera-Losada et al. (2009) y Ferreiro-Domínguez et al. (2011), en estudios realizados en la misma región, también encontraron que el número de especies y la proporción de las especies en el pastizal fueron modificadas por las prácticas de manejo, como la fertilización orgánica y el encalado. Sin embargo, en cualquier caso, es importante que estas prácticas de manejo sean compatibles con la conservación de las especies, que son características de los suelos ácidos de la zona atlántica, para mantener la biodiversidad vegetal.

CONCLUSIONES

I.-Más del 90% de los suelos gallegos son adecuados para recibir fertilizantes de lodos de depuradora bajo la actual norma RD 1310/90, pero sólo el 28,7% cumple con los requisitos de la EUDWD. La mayoría de las muestras que no cumplen con la regulación actual española están asociadas a rocas básicas y ultrabásicas que definen ambientes naturales con plantas específicas y microorganismos del suelo ya adaptados a estos niveles de metales pesados. Con el fin de aplicar prácticas más sostenibles para la producción agrícola, se propone tener en cuenta los niveles medios de metales pesados del suelo para cada metal pesado tratando de no superar los niveles medios de los suelos derivados de los diferentes materiales de roca madre. Además, esta recomendación respetaría el ambiente original del suelo que actúa como hábitat para diferentes organismos, preservando la biodiversidad beta.

II.- Basándose en los escenarios planteados 86/278/EEC (EU, 1986) (transpuesta en España mediante RD 1310/90, BOE, 1990), EUDWD (EC, 2000) y S1 y S2 (EC, 2008), los lodos de depuradora compostados deben ser utilizados como enmiendas del suelo de materia orgánica, mientras que los lodos estabilizados mediante digestión anaeróbica y granulados deben ser usados como fertilizantes para reducir la posible contaminación del suelo. Las normativas de lodos de depuradora basadas en las concentraciones del metal pesado presente en mayor concentración en el lodo (Zn) deben tratar de alcanzar los valores medios de Zn en el suelo normalmente asociado a diferentes materiales de roca madre para asegurar la salud microbiana del suelo y niveles sostenibles de Zn en cultivos o pastizales. Si no se mejora la calidad de los lodos de depuradora, la aplicación de los mismos debe reducirse en frecuencia o dosis, permitiendo que los suelos obtengan beneficios de aumentar la materia orgánica del suelo sin cambios fuertes en el contenido de suelo de Zn.

III.- El uso en agricultura de enmiendas orgánicas, elaboradas a partir de residuos orgánicos biodegradables como son los lodos de depuradora urbana que se estabilizan con cal, es una buena opción con respecto a las prácticas de manejo convencionales en Galicia, como son el encalado y la fertilización mineral. Esto es debido a que las enmiendas orgánicas basadas en residuos orgánicos implican un reciclaje de nutrientes al mismo tiempo que mejoran la fertilidad del suelo. En este estudio, la mejora de la fertilidad del suelo asociada a la aplicación de enmiendas orgánicas basadas en residuos orgánicos biodegradables (como lodos de depuradora, biorresiduos, estiércoles, etc) incrementó la producción de pasto y modificó su biodiversidad, principalmente cuando se aplicaron altas dosis de enmienda orgánica. La biodiversidad del pasto fue más sensible a la variación de la fertilidad del suelo causada por las enmiendas orgánicas que la propia producción del pasto, y por lo tanto la composición botánica podría ser considerada como un buen bioindicador de los cambios en la fertilidad del suelo cuando se aplica este tipo de enmiendas orgánicas elaborados a partir de residuos orgánicos. Además, los resultados de suelo y pasto asociados a las enmiendas orgánicas fueron similares a los obtenidos cuando se llevaron a cabo las prácticas convencionales que se realizan en la zona. Por lo tanto, la sustitución parcial o total de cal por las enmiendas orgánicas, incluso parcialmente del abonado mineral, podría ser una alternativa viable para reducir el coste de producción en las explotaciones gallegos y el impacto ambiental de los residuos orgánicos y los fertilizantes químicos.

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PUBLICACIONES

ARTÍCULOS, PUBLICACIONES O DOCUMENTOS CIENTÍFICO-TÉCNICOS

(CLAVE: L = libro completo, CL = capítulo de libro, A = artículo, R = review, E = editor, S = documento científico-técnico restringido.)

- Autores (p.o. de firmas): Mosquera-Losada, R., Amador-García, A., Muñoz-Ferreiro, N., Santiago-Freijanes, J.J Ferreiro-Domínguez, N., Romero-Franco, R., Rigueiro-Rodríguez, A.,
  Título: Sustainable use of sewage sludge in acid soils within a circular economy perspective
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  Editorial (si libro): Lugar de publicación:
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- Autores (p.o. de firmas): Amador-García, A., Ferreiro-Domínguez, N., Rigueiro-Rodriguéz, A., Mosquera-Losada, R.,
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- Autores (p.o. de firmas): Mosquera, M.R., Amador, A., Rigueiro, A.,
  Título: Effect of type and dose of sewage sludge application in maize+ryegrass rotation in Galicia (NW Spain).
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  Título: Potential sewage sludge utilization to fertilize forage surfaces in Galicia
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- Autores: Mosquera-Losada M.R., Amador-García, A., Rigueiro-Rodríguez, A.
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- Autores: Rigueiro, A., Amador, A., Mosquera, M.R.
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Sustainable use of sewage sludge in acid soils within a circular economy perspective

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ABSTRACT

Fertiliser future shortage and the associated increased economic and environment transport costs are key reasons to promote the use of urban residues as fertilisers in agriculture. Sewage sludge (SS) use as a fertiliser is promoted by the EU, which also consider the harmful effects of heavy metals (HMs). It is important to characterise the levels of HM in soils allocated to pasture and forage crops, before SS application, in order to have an initial soil reference and to see how they vary over time specially in acid soils, where HM availability and therefore its ecosystem impact is larger. For this study, we selected a region with natural very acid soils, and with high needs of fertilisers due to the high crop production potential regarding to important crops like maize, forage crops and grasslands. Galicia is a small region of Spain (9% of the Spanish territory) that produces the 33% and the 60% of the woodland products of Spain. This study generally aims to evaluate the use of SS as a fertiliser in a large agronomic region of Spain, as well as to identify the disadvantages associated with its use. In concrete terms, the study aims at comparing the implications of the current limit values and the analysis of the implications of tighter limits imposed by the EC to evaluate the effect on the availability of sites for sludge applications in Galicia. Results indicate that, after the analysis of 2557 soils, more than the 90% of Galician soils are suitable to receive sewage sludge (SS) following the current regulation RD 1310/90 but only 28.7% fulfil the EUDWD (European Union Draft Working Document) requirements. Most of the samples that do not fulfil the Spanish regulation are associated with basic and ultrabasic rocks that define natural environments with specific plants and soil microorganisms already adapted to these levels of HM. In order to apply more sustainable practices for agricultural production, it is proposed to take into account the mean HM levels of the soil for each heavy metal (HM) trying not to surpass the mean levels of the soils derived from the different parent rock material, after considering human health risks. Moreover, this recommendation would respect the original environment of the soil that acts as a habitat for different organisms, preserving beta biodiversity.

1. Introduction

Soil sustainable use and health is one of the key aspects to maintain and promote sustainable agriculture production. Fertilization is one of the most spread management activity linked to agriculture. However, due to the economic and environment costs of the fertiliser transport, fertilization based on farm surrounding residues is promoted within the FAO mixed farming concept (FAO, 2015). Urban-agriculture exchange of energy and nutrients should be promoted through activities (i.e. kilometer zero) like the use of urban residues in agricultural lands.

The bioeconomy concept relies on addressing inter-connected societal challenges such as food security, natural resource scarcity, fossil resource dependence and climate change, while achieving sustainable economic growth. The bioeconomy concept provides a useful basis for such approach, as it encompasses the production of renewable biological resources and the conversion of these resources and waste streams into value added products, such as food, feed, bio-based products and bioenergy (EC, 2012). It is essential to add-value to waste products, of which SS is one of the most important ones due to the large amounts of production of this residue in Europe as already recognized the FAO Smart Climate Agriculture document (FAO, 2013).

High levels of contamination in continental waters caused by humans promoted the establishment of the waste water directive in the nineties in Europe. Waste water is being also used as a source of nutrients in many countries (Ghafoor et al., 2012). The production of municipal SS in Europe has been increased since the start of the 1990s of the last century due to the implementation of the 91/271/CEE Directive,

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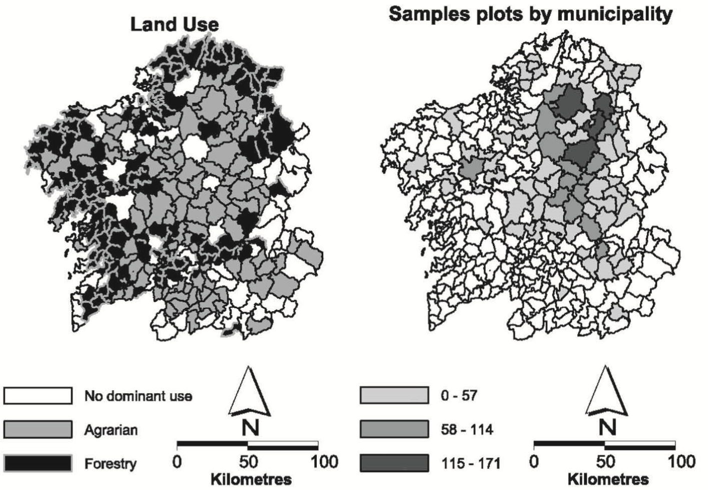


Fig. 1. Galician dominant land use (left) and number of soils sampled by county (right) in the current experiment.

which makes compulsory to treat continental waters in all cities with >2000 inhabitants. So, the availability of this residue is ensured and usually perceived as a challenge due to the high level of nutrients that can provide to crops.

Sewage sludge elimination could be done by transport to landfill, by incineration, which causes nitrogen release into the atmosphere (Smith, 1996; EPA, 2012) or by using it in soils. The use of SS as a fertiliser in soils is promoted by the European Union due to its nutrient contents, mainly nitrogen but also phosphorus, which could increase the value of this residue that otherwise, would generate environmental problems (Smith, 1996; 91/271/CEE Directive). This practice is also in line with the bioeconomy and circular economy concepts, providing an added value to the residue. One of the main concerns of SS use in soils as a fertiliser is related to the higher levels of HM and other organic substances compared with soils, which could be absorbed by plants and affect human beings through the food chain (Roy and McDonald, in press). Moreover,

Table 1
Statistical summary of selected properties for 2597 Galician soil samples used in this study compared with current and draft regulations. Min: minimum value, Max: maximum value; SD: standard deviation; notice that there were 1558 samples (41%) out of 2597 with pH below 5. Values between brackets represent the percentage of all samples for each directive.

	Value						
Property	Min–Max	Mean	SD	Median	Skewness	Kurtosis	90 percentile
pH-H_2O	3.44–10.22	5.21	0.72	5.17	1.25	5.67	6.05
Cd (mg kg^{-1})	0.01–2.50	0.05	0.15	0.01	5.63	50.07	0.10
Ni (mg kg^{-1})	0.001–169.50	12.20	14.28	8.90	2.31	10.80	27.70
Pb (mg kg^{-1})	0.01–118.50	10.21	12.13	6.60	1.75	5.34	27.12
Zn (mg kg^{-1})	0.01–306.70	45.33	30.46	41.20	1.58	6.39	81.92
Hg (mg kg^{-1})	0.01–0.80	0.054	0.05	0.05	5.33	52.08	0.10
Cr (mg kg^{-1})	0.01–236.94	10.03	16.54	5.80	4.87	37.09	22.32
Cu (mg kg^{-1})	0.01–212.00	16.38	17.98	12.30	3.73	23.98	34.70

Property	Value																	
	Legal requirements for use sewage sludge in soils. Samples below/above the limits																	
	RD 1310/1990 (*Directive 91/271/EEC*)						3rd Draft Working Document on sludge (EU)											
	pH ≤ 7	n = 2559		pH > 7	n = 38		5 ≤ pH < 6	n = 1230		6 ≤ pH < 7	n = 263		pH > 7	n = 38				
	Limit	Below	Over	Limit	Below	Over	Limit	Below	Over	Limit	Below	Over	Limit	Below	Over			
Cd (mg kg^{-1})	1	2557 (98.4)	2 (0.1)	3	38 (1.5)	0 (0)	0.5	1203 (46.3)	27 (1.0)	1	262 (10.1)	1 (0)	1.5	38 (1.5)	0 (0)			
Ni (mg kg^{-1})	30	2408 (92.7)	151 (5.8)	112	38 (1.5)	0 (0)	15	770 (29.6)	460 (17.7)	50	261 (10.1)	2 (0.1)	70	36 (1.4)	2 (0.1)			
Pb (mg kg^{-1})	50	2548 (98.1)	11 (0.4)	300	38 (1.5)	0 (0)	70	1230 (47.4)	0 (0)	70	263 (10.1)	0 (0)	100	38 (1.5)	0 (0)			
Zn (mg kg^{-1})	150	2540 (97.8)	19 (0.7)	450	38 (1.5)	0 (0)	60	880 (33.9)	350 (13.5)	150	262 (10.1)	1 (0)	200	37 (1.4)	1 (0)			
Hg (mg kg^{-1})	1	2559 (98.5)	0 (0)	1.5	38 (1.5)	0 (0)	0.1	1090 (42.0)	140 (5.4)	0.5	263 (10.1)	0 (0)	1	38 (1.5)	0 (0)			
Cr (mg kg^{-1})	100	2550 (98.1)	9 (0.3)	150	38 (1.5)	0 (0)	30	1111 (42.8)	119 (4.6)	60	263 (10.1)	4 (0.2)	100	38 (1.5)	0 (0)			
Cu (mg kg^{-1})	50	2504 (96.4)	55 (2.1)	210	38 (1.5)	0 (0)	20	837 (32.2)	393 (15.1)	50	263 (10.1)	0 (0)	100	38 (1.5)	0 (0)			

Table 2
t Student comparison between samples that do, or do not fulfil the Spanish regulation RD 1310/90.

	Samples that fulfil the Spanish regulation RD 1310/90		Samples that not fulfil the Spanish regulation RD 1310/90		
	Mean	EE	Mean	EE	*t*
Cd (mg kg^{-1})	0.05	0.003	0.05	0.02	−0.79
Ni (mg kg^{-1})	9.64	0.19	47.05	1.57	−23.70***
Pb (mg kg^{-1})	9.77	0.23	14.90	1.46	−3.46***
Zn (mg kg^{-1})	41.93	0.52	88.82	3.47	−13.36***
Hg (mg kg^{-1})	0.05	0.001	0.05	0.004	1.26
Cr (mg kg^{-1})	8.95	0.28	25.07	2.61	−6.14***
Cu (mg kg^{-1})	13.63	0.23	52.63	3.00	−12.95***

*** $p < 0.001$.

the use of SS as fertiliser could cause important soil degradation, including physical, chemical and microbiological damages (Yang et al., 2012), which could have a short, medium and long term impact and therefore affecting soil health (Fernández-Calviño et al., 2013).

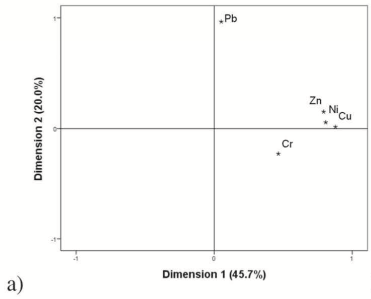
The European Directive 91/271/EEC which has the same HM thresholds as the Spanish Royal Decree 1310/1990 established limits for the use of SS as a fertiliser mainly by focusing on HMs. Moreover, the European Union launched a draft of the Directive to reduce even further the limits of allowable SS use in agriculture, on the European Union scale, in 2000 ((EC, 2000) (*Brussels, 27 April 2000 - ENV. E.3/LM*)). The so called 3 draft working document on sludge produced by the EU (EUDWD (EC, 2000)) was not approved due to the lack of consensus between the different countries but it could give us an idea of the next steps that will be taken in the regulation of the use of SS in agriculture at a European level and how to become more eco-efficient. This confirms that it is not easy to establish more strict limits at European level, and that other approaches are needed. The first step to reduce the impact of SS use is to monitor soils previous SS inputs and control how the soil evolution will take place (Vacca et al., 2012).

The HM concentrations of the SS, of the soils where it would be applied and the maximum quantity of HMs that could be applied in a 10 year period are currently limited by the approved regulations. The limits also depend on the soil pH, as HM availability increases as soil acidity is raised (Kabata-Pendias, 2001; Parat et al., 2005; Smith, 2009). Taking into account these directives, it is important to know what the base concentrations of HMs in municipal SS are, in different types of SS, that have already been evaluated at the Spanish national level (Mosquera-Losada et al., 2010). The second aspect to be evaluated is the level of HMs that already exist in the soils prior to adding SS, in order to study their capacity to receive SS as fertiliser under current and future directives, ensuring soil sustainability also for soil microorganisms (Guiller et al., 2009).

Galicia is a region with a large surface allocated to forestry (above 70% and producing 60% of wood in Spain (data from 2009)) (INE, 2012) but this Spanish region also produces >33% of the Spanish milk supply (data from 2008) mainly based on forage (crops like maize) and grasslands (INE, 2012). The use of SS on forested areas in low doses (50–100 kg of total N ha^{-1}) has been successfully tested in the north of Spain (Egiarte et al., 2009; Mosquera-Losada et al., 2011a) as well as in higher doses (160 kg of total N ha^{-1}) on grasslands (Ferreiro-Domínguez et al., 2011; Mosquera-Losada et al., 2011a, 2011b; Rigueiro-Rodríguez et al., 2000b, 2010a, 2010b, 2012). The use of SS on these types of land reduces the possible negative impact of HMs on human beings, as there is no direct consumption of the plants that may take up the HMs from the soil. Currently, >65% of SS produced in the region is used as fertiliser. Galicia soils could be considered representative of a large part of the Atlantic biogeographic region of Europe (EC, 2005; EEA, 2003). This study generally aims to evaluate the use of SS as a fertiliser in a large agronomic region of Spain, as well as to identify the disadvantages associated with its use. In concrete terms, the study aims at comparing the implications of the current limit values and the analysis of the implications of tighter limits imposed by the EC to evaluate the effect on the availability of sites for sludge applications in Galicia.

2. Materials and methods

The study was carried out in Galicia, a region found in the northwest of Spain, which occupies around 3 million ha. It is within the southwest part of the Atlantic biogeographic region of Europe. From 2007 to 2010, 2597 soil samples, which were never previously fertilised with SS, were taken randomly from privately owned plots in order to see whether they were suitable to receive SS. Soil samples were taken mostly from the agrarian based counties of the Galician region, where more fertiliser is needed compared with the predominantly forested counties (Fig. 1). The main agrarian activity in Galicia is related to forage crops and pasture to feed dairy and meat cows (IGE, 2011). The main forest species are *Pinus pinaster* Ait. and *Eucalyptus globulus* Labill, followed by a mixture of each of these two species with *Quercus robur* L. Soil was sampled at a depth of 25 cm as established by the Spanish Royal Decree 1310/90. Each sample was taken following the procedure indicated by the Spanish Royal Decree 1310/90: "a representative sample of soil per plot submitted to analysis will be composed of a mixture of 25 samples taken from an area of 5 hectares or less per plot". The number of hectares of the sampled plots represents approximately 20% and 1.5% of grasslands and agrarian soils (including forage crops and grasslands) of Galicia, respectively. Once taken, all soil samples were transported to the laboratory and air dried. Afterwards, soil samples were sieved through a 2 mm sieve. Later on, Ni, Cd, Zn, Cr, Cu and Pb concentrations were analysed with the VARIAN 220FS spectrophotometer using atomic absorption (VARIAN, 1989), after a nitric acid digestion made in a CEM



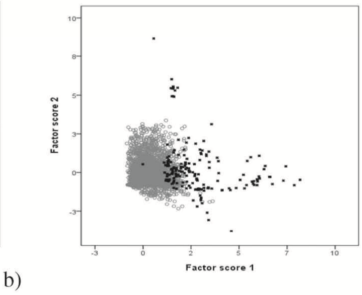


Fig. 2. Factorial analysis of heavy metals of all studied samples (panel a) and score factor plot of samples (panel b). Dark spots represent those soils with limits above the Spanish regulation. Varimax rotation, KMO = 0.69.

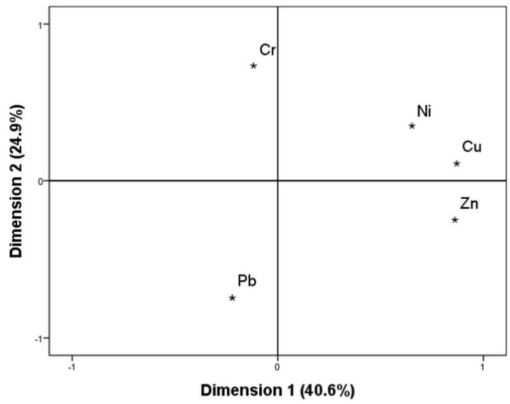


Fig. 3. Factor analysis of heavy metals with samples that do not fulfil the Spanish RD 1310/90. Varimax rotation KMO = 0.56.

MDS-2000 microwave (CEM, 1994), with Hg determined with hydride generator. Water soil pH was also measured (2.5:1) (Guitián and Carballas, 1976).

Descriptive statistics (mean, median, kurtosis, etc.) of the HMs were performed on the 2597 samples in the experiment and counting the number of samples that meet or fail legal requirements for SS use in agriculture. The descriptive statistics include median absolute deviation (mad), which is a robust method for evaluating dispersion. The *t* Student and *U* Mann Whitney tests were used for comparison of HMs between samples that fulfil and those samples that do not fulfil the Spanish regulations. The obtained results with both tests were similar due to the large sample size in this comparison that tended to diminish the detrimental effects of non-normality. Factor analysis and cluster analysis were used to evaluate the relationships between the concentrations of different HMs in those soils that did not fulfil the current regulations. Using only the metals with significant differences between the samples that meet and fail the current legal requirements, a factor analysis from an exploratory perspective was performed. Factor analysis provides two outcomes; data summarisation and data reduction (Hair et al., 2010). In summarising the data, factor analysis derives the underlying dimensions that, when interpreted and understood, describe the data in a much smaller number of concepts than the original individual variables (HMs). Data reduction extends this process by deriving an empirical value (factor score) for each dimension (factor) and then substituting this value for the original values. The purpose is to simplify the subsequent multivariate analyses.

The analyses of the similarities and differences among those plots that do not fulfil the Spanish regulations were performed with a dendogram derived from a cluster analysis applied to the standardised HMs data. Proximities between samples were calculated based on the squared Euclidean distance and Ward's method was used as linkage procedure. The cluster analysis is complementary to the factor analysis. Factor analysis makes groupings based on the patterns of variation (correlation) in the data, whereas cluster analysis makes groupings on the basis of distance (proximity) (Hair et al., 2010). Statistical calculations were performed using SPSS (PASW 18.0) for windows.

3. Results

Table 1 shows the results of the descriptive statistics of soil pH and HMs obtained from the 2597 samples taken in this experiment. Zn was the regulated HM with the highest concentration in the soil (45.33 mg kg^{-1}), followed by Cu, Ni, Pb and Cr with the lowest concentrations associated to Cd and Hg. The statistical parameters show that over 50% of the data are below the mean with skewness higher for the lowest mean values, such as Hg, and Cd and always positive, which means that most of the rare cases are associated to high values. All kurtosis values are positive, which is indicative of a higher concentration of values around the mean when compared with the normal Gauss curve.

3.1. Soil analyses vs. regulations

All HM mean values were below the thresholds indicated by the Spanish Royal Decree 1310/90 and the most restrictive limits described by the EUDWD (European Union Draft Working Document (EC, 2000)). If each HM is independently evaluated, 90% of the samples were always below the thresholds indicated for acid soils by the Spanish Royal Decree 1310/1990. However, all the maximum values for the evaluated soils were above the thresholds with the exception of the Hg, when compared with the values provided by the Spanish Royal decree for acid soils. Only 151, 55, 19, 11, 9 and 2 samples of our 2597 total samples were over the limits for Ni, Cu, Zn, Pb, Cr and Cd stipulated by the Spanish regulations, which represents a percentage of around 5.8%, 2.1%, 0.7%, 0.4%, 0.3% and 0.1% of all evaluated samples, when each HM is independently taken into account, respectively. If the EUDWD draft limits are considered, then around 41.0% of the samples could not receive SS, due to their pH level being lower than 5, irrespective of the HM concentrations. From the 1066 samples with pH levels below 5, 473 samples meet the limits set by the EUDWD draft regarding pH (between 5 and 6). Out of the remaining 59.0% of samples with pH above 5 and if each HM is individually taken into account, the number of soils that could not receive SS was 28, 464, 352, 140, 123 and 393 for Cd, Ni, Zn, Hg, Cr, and Cu, respectively. This makes a total of 1.8%, 30.3%, 22.9%, 9.1%, 8.0% and 25.7% of the soil samples with respect to the samples with pH above 5 and a 42.1%, 58.9%, 54.6%, 46.4%, 45.8% and 56.2% of the total number of samples (including those samples with pH below 5) that will not be able to receive SS as fertiliser for each respective HM.

When all limits are considered together, around 93.3% of samples (2424) meet the 1310/90 Spanish regulation (based on the Spanish implementation of the currently approved EU regulation - *Council Directive 86/278/EEC of 12 June 1986 on the protection of the environment, and in particular of the soil, when SS is used in agriculture*) and therefore 6.7% (173 samples) did not fulfil the Spanish regulations. Around 42.7% of the soils that do not fulfil the current regulations (173 out of 245

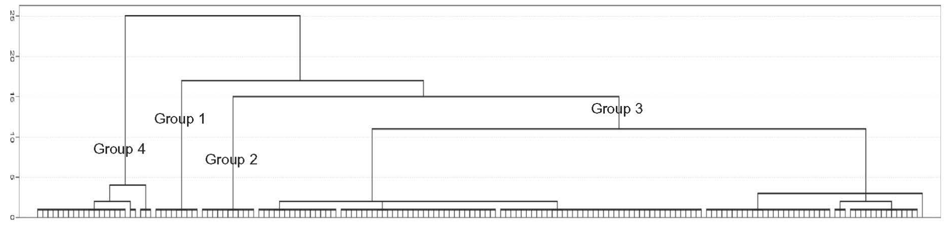


Fig. 4. Dendogram performed from cluster analysis applied to the standardised data of those plots that do not fulfil the Spanish regulation.

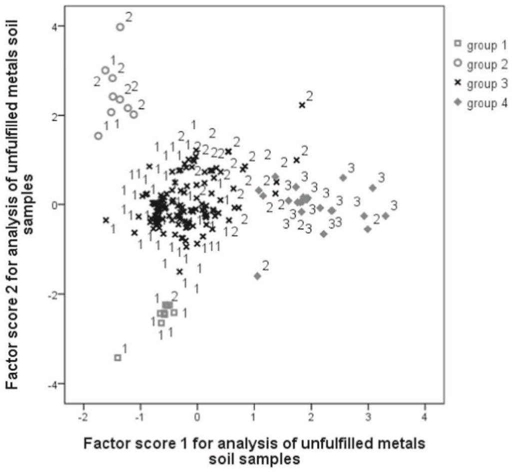


Fig. 5. Score factor plot for analysis of soil samples that did not fulfil heavy metals. Overprinted numbers represent the number of metals that the sample does not fulfil.

determinations) do so for >1 HM and this makes it advisable to evaluate the relationship between the concentrations of different HMs, when trying to find an underlying factor that affects them. When the EUDWD is taken into account, only 28.7% of soils could receive SS. For each pH interval, the samples that could not receive SS were 41.0%, 63.0%, 2.6% and 5.3% for pH below five, 5–6, 6–7 and above 7, respectively. If each determination is evaluated, 1500 samples will not be able to receive SS, which means around 91% (1500/784) of the samples did not fulfil the EUDWD because of more than one HM, if only samples with pH above 5 are considered.

3.2. Heavy metal soil relationship in soils that do not fulfil Spanish regulations

When a comparison of each HM between the soil groups that fulfil and do not fulfil the Spanish regulation RD 1310/90 was performed, it was found that they were all significantly different with the exception of Cd and Hg (Table 2). Differences between those soils that fulfil the Spanish regulation RD 1310/90 were: one and a half-fold for Pb, two-fold for Zn, 3-fold for Cr, 4-fold for Cu and 5-fold for Ni.

However, the Box-Whisker figures (not shown) of each HM reveal that there is an overlapping between those soils that fulfil and those that do not fulfil the Spanish regulation. Thus, it is not possible to discriminate between the soils that are able to receive SS under the current regulations, based on any particular HM. Therefore, a multivariate approach was performed. A factor analysis (with principal components extraction, followed by VARIMAX rotation) was used to investigate whether the HMs with significant differences (Ni, Pb, Zn, Cr and Cu) represent identifiable underlying factors in those samples with soil pH lower than 7. Factor analysis (Fig. 2a) showed that total soil concentrations of Cu, Ni and Zn are related and described by the factor 1 with Pb described by factor 2. This means that those samples with high levels of Zn also have high levels of Ni and Cu and they are independent of samples with high concentrations of Pb. Fig. 2b shows that those samples that do not fulfil the Spanish regulations have high values for the scores of factor 1 and/or factor 2.

On the other hand, there are 40.1% of samples that have more than one HM that does not fulfil the Spanish regulations to receive SS. Out of 173 soils that do not fulfil the Spanish regulations, there are 116, 40 and 17 samples that have one, two or three HMs that exceed the values stipulated by the Spanish regulations, respectively. Fig. 3 shows the results of a factorial analysis carried out for the concentrations of Cr, Pb, Ni, Cu and Zn within those samples that do not fulfil the RD 1310/90 regulation. The total soil concentrations of Ni, Cu and Zn are related to Factor 1, which means that those samples with high values of Ni also had high values of Cu and Zn. Factor 2 is positively and negatively highly correlated with the levels of Cr and Pb, respectively. This fact makes it advisable to carry out a hierarchical cluster analysis to draw the dendogram described in Fig. 4.

The cluster analysis allows us to divide those samples that did not fulfil the current regulations into groups. At the indicated similarity level of 12, a division of samples into four groups was chosen; the two smaller groups (group 1, group 2) consisting of 9 and 11 soil samples, respectively. The next largest group is formed by 130 soil samples and the remaining group is composed of 23 soil samples, being groups 3 and 4, respectively. The groups were based on the metal that exceeds the Spanish regulation limits and the number of HMs that the soil samples have above those limits.

Fig. 5 shows the scores reached by the soils that do not fulfil the Spanish regulations for both factors extracted as explained in Fig. 3. Fig. 4 and Table 3 show that groups 1 and 2 are those samples that have highest Cr and Pb values, respectively. Soil groups 3 and 4 are those that have levels of Ni, Zn or Cu higher than the specifications given by the Spanish Royal Decree; particularly most of the soil samples of Group 4, which have the highest levels of factor 1 because they exceed the levels for three HMs at the same time. Within group 3, the samples with the highest scores are those that have both Ni and Cu as the HMs that exceed the limits, the rest of the samples of this third group did not fulfil the SS regulations for one HM, mostly Ni.

A description of the location of those samples that exceed in 1, 2 and 3 metals in each county and the percentage of soil samples per county that do not fulfil the Spanish regulations can be seen in Fig. 6. As was previously indicated, Ni is the HM that limits the use of SS in the highest number of plots per county and for this reason Ni limited the use of SS in a higher number of counties than the rest of the HMs. Pb, Cu, Zn and Cr limits the use of SS based on RD 1310/90 in some specific counties, such as Santiago de Compostela or Moeche (NW of Galicia), which are also the areas that have plots that do not fulfil the requirements of RD 1310/90 in respect of two or three HMs.

Table 3
Number of samples and heavy metals above limits in each group of unfulfilled metals soil samples.

	Metals above limits in samples into the group (no. of samples in branches)			
	Group 1	Group 2	Group 3	Group 4
Number of metals above limits				
1	Cr (3)	Pb (9)	Cd (2) Ni (94) Cu (8)	
2	Cr, Ni (6)	Pb, Ni (2)	Ni, Cu (26)	Ni, Zn (2) Ni, Cu (4)
3				Ni, Zn, Cu (17)
No. of samples in the group	9	11	130	23

4. Discussion

Soil HM ranges found in this study were within those generally described for Cd (Page et al., 1981) (0.01–3 mg kg^{-1}), Pb (Pais and Jones, 1997) (3–189 mg kg^{-1}), Cr (Alloway, 1995) (0.3–10,000 mg kg^{-1}) and Hg (Pais and Jones, 1997) (0.01–1.8 mg kg^{-1}). However, the essential elements for plants like Ni (Alloway, 1995) (1–100 mg kg^{-1}), Cu (Domínguez-Vivancos, 1997) (3–100 mg kg^{-1}) and Zn (Barber, 1995) (10–300 mg kg^{-1}) have a broader range than those values given in the literature for soils. The association of high levels of HMs with soils derived from ultrabasic and basic rocks soil parent material (Ross, 1994; Díez-Lázaro et al., 2002; Candeias et al., 2011) could explain the broader range of Ni and Zn as the factorial analyses carried out in our study demonstrate.

Our results showed that >90% Galician soils are suitable to receive SS under the current regulations. However, the fact that all mean and median values of the agrarian soils of the present study were below the HM baseline levels of all types of Galician soils (Macías-Vázquez and Calvo

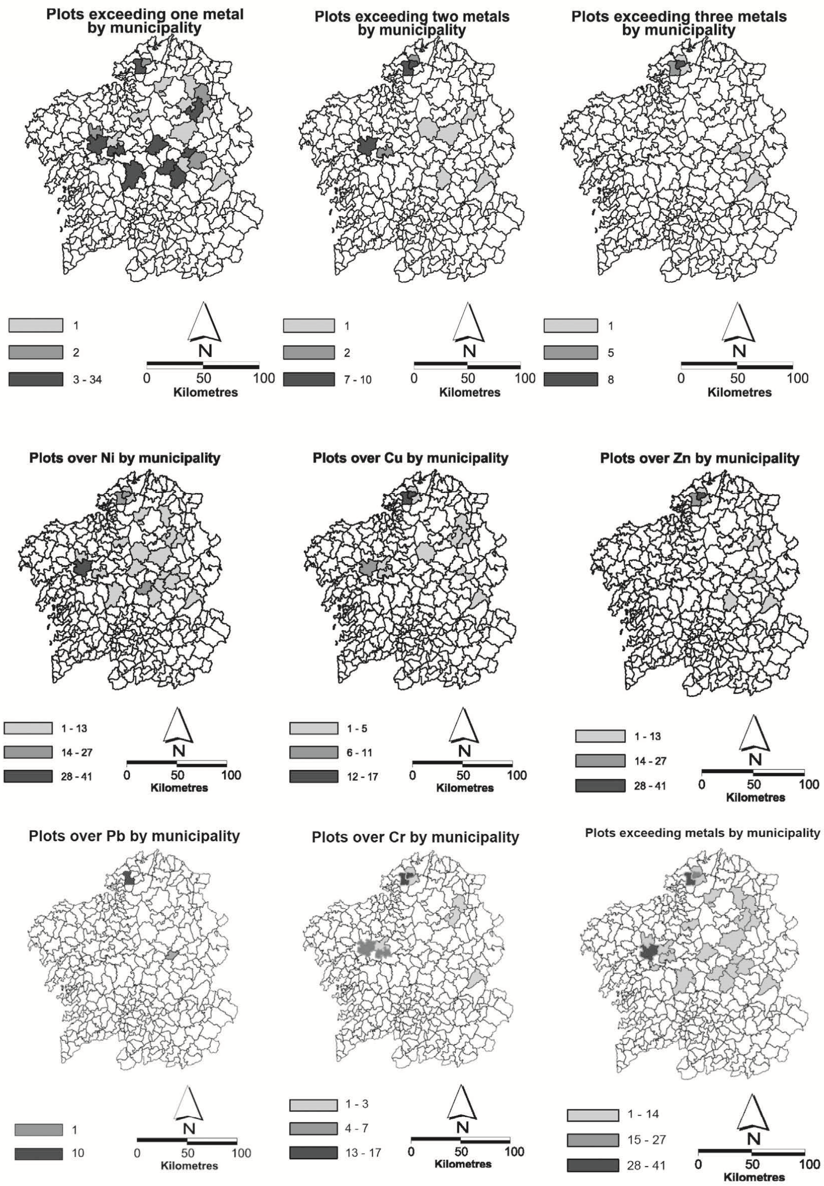


Fig. 6. Counties with plots that exceed the limits established by the RD 1310/90 in one, two and three heavy metals in Galicia.

de Anta, 2009), indicates that most of the Galician soils are suitable to receive SS fertilisation without a significant increase of the usual levels of HMs in soils for each type of rock parent material and if acceptable doses of SS with adequate quality (low HM levels) are applied in order to fulfil crop requirements. Recent revisions of the benefits of using high quality SS as fertiliser are provided by Diacono and Montemurro (2010), Lu et al. (2012) and Smith (2009).

Considering the mean chemical characteristics of the SS in Spain (Mosquera-Losada et al., 2010) stabilised by anaerobic digestion, we can estimate the amount of SS that should be applied in a soil for a given N rate (EPA, 1994). Once known the loading rate and the HM concentrations of SS, we can estimate the real inputs of HM per application and the minimum number of applications that SS could be used to reach the maximum concentrations in soil of HMs allowed by the Royal Decree 1310/90 in acid soils, assuming no relevant leaching or crop extractions of HMs from soils (McGrath, 1987). Anaerobic SS inputs of 200 kg of total N ha^{-1} (EPA, 1994) in a soil, implies a total increase of 1.26, 0.37, 0.05, 0.01, 0.16, 0.001 and 0.03 mg of Zn, Cu, Ni, Cd, Pb, Hg and Cr per kg^{-1} of soil and application, respectively (standard soil density of: 1.1 Mg m^3 and a depth of 0.25 m (RD 1310/90)). These figures allow a minimum number of applications of 82, 89, 304, 94, 253, 491 and 2474 (usually years because SS is annually applied) for Zn, Cu, Ni, Cd, Pb, Hg and Cr in a mean soil, respectively. Therefore, the essential nutrients Zn, Ni and Cu are the most limiting for applying SS, as found McGrath (1987) in soils evaluated after long term SS inputs.

Among all soil parameters that could modify soil HM fates, only soil pH is currently included in most of the regulations for SS use in agriculture. Soil pH directly affects HM availability (Parat et al., 2005; Lu et al., 2012) and therefore their fate: mainly leaching or crop extraction. It is known that most HMs precipitate in neutral and basic soils (McGrath, 1987), and only soil movement (i.e. ploughing) could explain HM depletion (McGrath, 1987). On the contrary, in acid soils and soils with a gradual reduction of organic matter, an effect called "sludge time bomb" can appear. Sludge time bomb means that more soluble forms of HM can be released from soils as organic matter mineralization happens and, later on, taken by crops. This could explain, for example, why soil total bacterial populations are initially improved after nutrient supplies with SS inputs and later depleted when HMs availability is increased in soils (Guiller et al., 2009). The "sludge time bomb" concept, that could be more associated to acid soils, makes important to propose criteria for sludge inputs in soil ensuring that mean soil values of HMs for each type of soil are not surpassing. Having soil mean values for each type of parent rock material as a criterion for applying SS probably ensures that microbial and natural plant population can survive in those already existing environments, therefore promoting sustainability and preserving beta biodiversity among different types of soil conditions. Moreover, preserving soil mean values of HMs for each type of rock parent material, will also be adequate for sustainability of agricultural production of human food, as these soils were usually used for crop production.

The current EU and Spanish regulations are based on pH at a broad range. Most of the Galician soils have an acid pH, as happen with the agrarian soils evaluated in this study (only 1.5% of the soil samples have a pH above 7). The EUDWD draft does not mention the levels of HMs below which the sludge could be applied if pH is below 5, which directly affects 41% of the Galician soils. Even though, soil acid pH is a key factor increasing soil HM solubility (Parat et al., 2005) and therefore its potential to be leached and uptaken by crops, pH indicator has several concerns as a tool for SS applications for acid soils. Natural Galician soils are generally below pH 5 when no lime is applied, mainly due to the rainfall regime and crop extraction. Recommendations for applications of lime every four years in Galicia are usually based on the reduction of the percentage of saturation of Al to below 20%, which can be frequently obtained if the soil pH is around 5.5. If soil pH is artificially increased with lime applications, SS inputs are allowed and therefore the "sludge time bomb" process could be easily started, once liming is ceased. Of the soils analysed in the current study, 41 and 88% had a pH below 5 and 6, respectively. This means that if SS was used because soil pH is above 5 and no lime was applied for a long period of time, then soil pH could be easily reduced. On the other hand, even though most HMs are more available at low soil pH, some such as Cu could become less available below 5 than over this value (Porta, 2010). Therefore, it may be better to take into account the existing baseline levels of HMs in soils and to control the maximum dose permitted to be added as a percentage of that existing baseline levels. This will allow the application of more targeted amounts of SS, depending on the type of soil and the soil parent material, avoiding significant increases in soil HMs in a controlled way, which will protect the ecosystem at the same time, and probably reduce the negative long term impacts of SS inputs on soil total and nitrogen fixing microorganisms (Guiller et al., 2009). Baseline mean levels for each type of soil will probably allow the application of SS in >44.37% (473/1066) of soils with pH below 5, as all the mean HMs are below the lowest thresholds established by the EUDWD draft regulation. On the other hand, the reduction in sludge application advised by EUDWD has avoided the necessity to implement the EUDWD draft regulation. Moreover, it is also important to recommend other practices, such as the use of high quality sludge, the incorporation of SS in soils with ploughing to avoid a gradient of SS and to forbid direct grazing after applications, as animals consume large amounts of soil (ranging between 182 and 803 kg per year in dairy cows (Herlin and Andersson, 1996)) and therefore of HMs. The frequency of application on the same land should also be reduced.

5. Conclusions

Soil HM ranges found in this study were within those generally described for Cd, Pb, Cr and Hg, with the exception of Ni, Cu and Zn that have a broader range than those values given in the literature for soils. The high levels of Ni, Cu and Zn are explained by the fact that some soils derived from ultrabasic and basic rocks soil parent material. More than 90% of Galician soils are suitable to receive SS fertiliser under the current regulation RD 1310/90, but only 28.7% fulfil the EUDWD requirements. Most of the samples that do not fulfil the Spanish current regulation are associated to basic and ultrabasic rocks that define natural environments with specific plants and soil microorganisms already adapted to these levels of HMs. In order to apply more sustainable practices for agricultural production, it is proposed to take into account the mean HM levels of the soil for each HM trying not to surpass the mean levels of the soils derived from the different parent rock material, after considering human health risks. Moreover, this recommendation would respect the original environment of the soil that acts as a habitat for different organisms, preserving beta biodiversity.

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